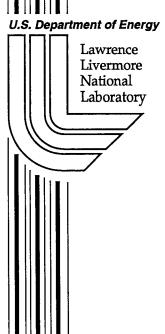
# Evaluation of the Radiochemistry of Near-Field Water Samples at the Nevada Test Site Applied to the Definition of a Hydrologic Source Team

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# EVALUATION OF THE RADIOCHEMISTRY OF NEAR-FIELD WATER SAMPLES AT THE NEVADA TEST SITE APPLIED TO THE DEFINITION OF A HYDROLOGIC SOURCE TERM

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### **EXECUTIVE SUMMARY**

For almost three decades radiochemical data have been collected from water samples that originate from saturated nuclear test cavities, chimney rubble zones, and satellite wells constructed adjacent to underground nuclear tests conducted on the Nevada Test Site (NTS). Although these data have been collected regularly under the auspices of Defense Programs and Environmental Management funded programs, there has been less effort made at interpreting the data to define controls on the dissolved or mobile fraction of radionuclides in groundwater. The need for data analysis have been given more impetus by the recent effort to construct sophisticated models of hydrologic flow and contaminant transport in the near-field. These models rely on input parameters and boundary conditions which can best be obtained through field observations of radionuclide migration adjacent to nuclear tests. The present report is an attempt to synthesize observations and analyze data returned from the near-field over the past ten years and draw general conclusions regarding the behavior of radionuclides in Nevada Test Site groundwaters.

The complexity of the hot well problem deserves mention. Most nuclear tests were conducted between ~ 500 and ~ 1200 m below the surface. Besides the obvious residual radioactivity, the post-test environment is characterized by extreme heterogeneity due to the mechanical damage done to the rock units at the working (firing) point. For these reasons, the construction of wells and collection of water samples is a technically complex, expensive, and logistically complicated enterprise. While data exists, it must be interpreted with care and, for these reasons, data interpretations have been limited in scope. In the majority of cases, near-field or "hot" wells were completed opportunistically in post-shot re-entry borings into a nuclear test cavity or chimney, originally drilled to return solid samples diagnostic of device performance. Of the more than eight hundred nuclear tests conducted at the NTS, only fifteen near-field study sites regularly produce water. It must be emphasized that the near-field or "hot well" sampling program is not a monitoring program designed to detect the arrival or movement of radionuclides. Rather the data from these wells can be used to establish bounds on those radionuclides in solution and to deduce controls on their solubility and mobility. Data returned from near-field wells are affected by well completion techniques and in particular by whether the water is pumped or bailed. Only pumped samples are truly representative of water in the sampling interval, and time-series analyses of pumped cavity water samples have proven particularly important in illuminating the nature of radionuclide migration.

The justification for a program to collect near-field groundwater samples can be capsuled in five points. First, hot well sampling and analysis provides an the empirical measure of radionuclide solubility and resulting concentrations of importance to determining risk to human health and the environment. Second, analytical methodologies developed in the past five years afford a unique opportunity to measure the hydrologic source term at extremely low concentrations. <sup>3</sup>H, <sup>14</sup>C, <sup>36</sup>Cl, <sup>85</sup>Kr, <sup>90</sup>Sr, <sup>99</sup>Tc, <sup>129</sup>I, <sup>137</sup>Cs, <sup>235</sup>U, <sup>236</sup>U, <sup>237</sup>Np, <sup>239</sup>Pu, <sup>240</sup>Pu, and <sup>241</sup>Am may be measured at concentrations less than 0.05 Bq/L with high precision and high abundance sensitivity. Third, data from near-field wells not

only contains information on radionuclides in solution but also may be utilized to determine the physiochemical conditions of the water matrix. These data are a necessary compliment to the radioinuclide data, and have direct bearing on the rate of radionuclide release from solid matrices. Fourth, regular sampling of hotwells provides necessary experience in collecting radiologically contaminated samples, assessing their quality, trending the results, and establishing a radiological baseline that services both operational and regulatory needs. Finally, hot well data provide the only means to validate and verify numerical models of radionuclide concentrations in the near-field. For these reasons, a regular effort to collect and analyze these data is an essential component of a comprehensive strategy to assess radionuclide migration at the NTS. Data from the Nevada Test Site also provides valuable insight into radionuclide migration under ambient groundwater flow conditions that can be used at other sites – particularly those associated with nuclear waste disposal – to better understand the potential for radionuclide mobility.

A primary conclusion of this summary is that widespread contamination of groundwater aquifers at the Nevada Test Site is unlikely. Relative to the radiologic source term, only a small fraction of the actinides, fission products, and activation products are measured in near-field groundwaters. A majority of the tests were conducted above the water table where radionuclides are assumed to unavailable for transport by groundwater. In addition, some tests appear to seal themselves, resulting in little interaction with the ambient groundwater regime. A majority of tests conducted in Pahute Mesa, Yucca Flat, and Frenchman Flat were detonated in zeolitized ash-flow and ash-fall tuffs or alluvial sediments that sorb radionuclides. Hydrologic gradients in the Frenchman Flat and Yucca Flat testing areas are low enough that lateral migration of radionuclides are extremely slow and will be governed by flow rates in the regional carbonate aquifer, should radionuclides reach these depths. Where groundwater flow is static even tritium is relatively immobile.

Other conclusions of data acquisition and analysis in the near-field include:

- Highly soluble radionuclides include <sup>3</sup>H, <sup>14</sup>C, <sup>36</sup>Cl, <sup>85</sup>Kr, <sup>99</sup>Tc, and <sup>129</sup>I. These radionuclides are found as dissolved species and scale proportionally to concentrations of tritium (i.e., a 50% tritium concentration equates to a 50% concentration of <sup>85</sup>Kr when scaled to the cavity concentration of that species for a given nuclear test). Ionic radionuclides including <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>239</sup>Pu although encountered in cavity and chimney waters are not transported readily outside the immediate cavity-chimney environment.
- Groundwater flow is required to mobilize even highly soluble radionuclides; for this reason there is a higher potential for lateral movement of radionuclides on Pahute Mesa than Yucca Flat or Frenchman Flat.
- Time-series analyses provide information of the control of radionuclide migration including dilution, dispersion, leaching and precipitation.
- Relatively insoluble radionuclides including plutonium may be transported at ambient groundwater velocities through sorption to clay, zeolite, and feldspar colloids suspended in fractured rock aquifers. Resulting concentrations are below drinking water standards.

• Residual heat from underground nuclear tests may help mobilize radionuclides through the ascent of soluble species to more transmissive aquifers. Other tests have little interaction with ambient groundwater and remain isolated for decades following their detonation.

 Prompt process may mobilize radionuclides along specific pathways that are related the rock strength and geologic structure; gas phase transport may cause volatile radionuclides to ascend and be deposited at a shallower

interval in a nuclear test chimney.

### INTRODUCTION

Effective management of available groundwater resources and strategies for remediation of water impacted by past nuclear testing practices depend on knowledge about the migration of radionuclides in groundwater away from the sites of the explosions. A primary concern is to assess the relative mobilities of the different radionuclide species found near sites of underground nuclear tests and to determine the concentration, extent, and speed of this movement. Ultimately the long term transport behavior of radionuclides with half-lives long enough that they will persist for decades, their interaction with groundwater, and the resulting flux of these contaminants is of paramount importance.

As part of a comprehensive approach to these assessments, more than three decades of site-specific sites studies have been undertaken at the Nevada Test Site (NTS) which have focused on the means responsible for the observed or suspected movement of radionuclides away from underground nuclear tests (RNM, 1983). More recently regional and local models of groundwater flow and radionuclide transport have been developed as part of a federal and state of Nevada program to assess the long-term effects of underground nuclear testing on human health and environment (e.g., U.S. DOE/NV, 1997a; Tompson et al., 1999; Pawloski et al., 2001). Necessary to these efforts is a reliable measure of the hydrologic source term which is defined as those radionuclides dissolved in or otherwise transported by groundwater (Smith et al., 1995).

Measurement of radionuclides in waters sampled near the sites of underground nuclear test provides arguably the best opportunity to bound the hydrologic source term. This empirical approach was recognized early and concentration data has been collected annually since mid-1970's. Initially three sites were studied at the NTS; over the years the program has been expanded to include more than fifteen study locations.

As part of various field programs, Lawrence Livermore National Laboartory and Los Alamos National Laboratory have annually returned water samples from wells in near-field locations at the NTS for radiochemical analyses. This report makes the distinction between samples taken in the near-field and the far field. The near-field includes the area extending radially ~ 300 meters from surface ground zero (the firing point of an underground nuclear test projected upwards on ground surface). Over the years this sampling program has also been refereed to as the "hot-well monitoring program" because these water samples contained concentrations of tritium above natural background (tritium concentrations in southern Nevada precipitation are 0.5 to 2.0 Bq/L, Farmer et al., 1998). A majority of the hot wells contain tritium in excess of the 741 Bq/L (20,000 pCi/L) drinking water standard (Smith et al., 1996a; Smith et al., 1997). The sites which comprise our current hot well sampling network are plotted on a map of the NTS in Figure 1.

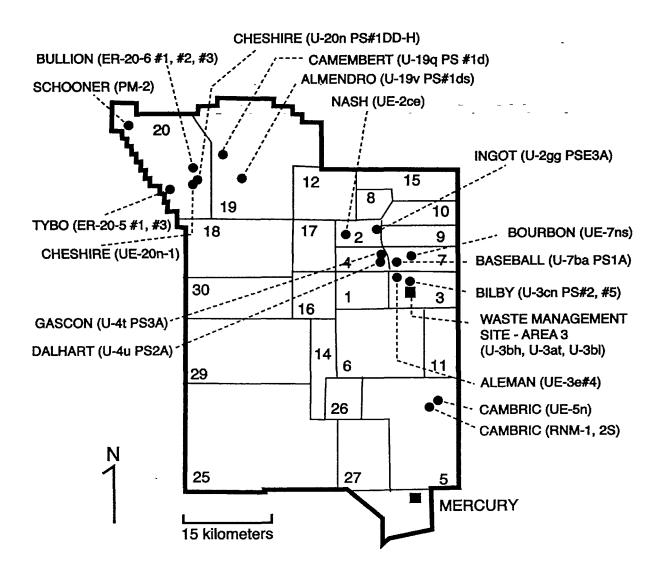


Figure 1. Map of the Nevada Test Site showing near-field sampling sites; both test name and affiliated near-field well (in parentheses) are listed.

Table 1 lists near-field sampling sites, test related data, and affiliated well name. Hole name, test date and announced yield from U.S. DOE/NV (2000); depth of burial and estimated water table (in meters) from U.S. DOE/NV (1997b).

TABLE 1
Near-Field Sampling Sites

Test	Hole	Well	Test Date	Yield (kilotons)	Depth of burial (m)	Depth to water (m)
ALEMAN	U-3kz	UE-3e#4	9/11/1986	<20 kt	503	475
ALMENDRO	U-19v	U-19v PS#1ds	6/6/1973	200 – 1000 kt	1067	707
BASEBALL	U-7ba	U-7ba PS1A	1/15/1981	20 – 150 kt	564	458
BILBY	U-3cn	U-3cn PS#2, U-3cn #5	9/13/1963	249 kt	715	509
BOURBON	U-7n	UE-7ns	1/20/1967	20-200 kt	560	601
BULLION	U-20bd	ER-20-6	6/13/1990	20-150 kt	674	621
CAMBRIC	U-5e	RNM-1, 2S, UE-5n	5/14/1965	0.750 kt	295	218
CAMEMBERT	U-19q	U-19q PS1D	6/26/1975	200-1000 kt	1312	666
CHESHIRE	U-20n	U-20n PS#1 DD-H; UE-20n-#1	2/14/1976	200-500 kt	1167	631
DALHART	U-4u	U-4u PS2A	10/13/1988	< 150 kt	640	477
GASCON	U-4t	U-4t PS3A	11/14/1986	20 150 kt	593	490
HYRAX	U-3bh	U-3bh	9/14/62	low	217	491
INGOT	U-2gg	U-2gg PSE3A	3/9/1989	20 – 150 kt	500	563
NASH	U-2ce	UE-2ce	1/19/1967	39kt	364	527
TYBO	U-20y	ER-20-5	5/14/1975	200 – 1000 kt	765	630

Sampling, measurement, and interpretation of radionuclides from the near-field environment has proven to be logistically complicated due to the relative inaccessibility of the test cavities created by the explosions, sampling artifacts introduced during the entry and collection of these waters, the limited number of sampling sites and relative infrequency of sampling operations (e.g., Smith et al., 1995).

At the request of the Environmental Management program of the U.S. Department of Energy, Nevada Operations Office (DOE/NV), radiochemical and trace element data collected by the national laboratories from near-field locations is compiled and reviewed with an emphasis on using this data to define a hydrologic source term for the Nevada Test Site. Although data collection activities have been on-going for decades, there has little effort to systematically analyze this data for use in the remedial effort or to justify additional data collection activities. In particular peer-reviewers and regulators have critiqued proposed recent remedial investigation plans because existing near-field data was not used to adequately support numerical predictions of contaminant flux (NDEP, 1999a; NDEP, 1999b; NDEP 1999c).

The purpose of this review is to provide a short history of near-field data collection operations at the Nevada Test Site, describe the hydrogeologic and radiochemical setting of near-field locations, assemble a representative set of data for comparison, assess data quality, review prior data analysis, and offer

conclusions regarding the application of this data to developing a hydrologic source term.

### HISTORY

Initial questions posed by the Nevada Test Site radionuclide migration program concerned how much of the radioactivity residual from a nuclear test site is available to groundwater system, whether radionuclides were deposited in regional aquifers, the velocity of groundwaters in the vicinity of the tests, and the interaction between groundwater flow and contaminant transport (RNM, 1983). As early as the mid-1970's, a field program was initiated to "gain some field data bearing on the question of the relation of the hydrologic source to the radiochemical source term. The theories, predictions, and laboratory leaching experiments have been summarized...but...there is little or no field data." (Borg et al., 1976).

Radionuclide migration field programs were first sponsored by the Radionuclide Migration Program (RNM) which was chartered by Defense Programs of the Department of Energy, Nevada Operations Office in 1974; the RNM evolved into the Hydrology/Radionuclide Migration Program (HRMP) in 1988 and then again into the current Hydrologic Resources Management Program in 1993 (U.S. DOE/NV, unpublished; U.S. DOE/NV, 1993). Since the end of underground nuclear testing in 1992, budgets for the HRMP were reduced while support for environmental remediation activities increased. The Groundwater Characterization Program (GCP) began at the NTS in 1991. GCP was renamed the Underground Test Area (UGTA) in the mid-1990's. UGTA field studies have included hydrologic characterization, 'nature and extent' near-field test wells, and field-scale radionuclide and tracer transport experiments. DOE/NV's current Routine Radiological Monitoring Program (RREMP) captured an array of existing NV funded long-term groundwater monitoring activities and relies on near-field data as part of a monitoring program that focuses on protection of water supply wells, permitted facilities, and aquifer monitoring.

Recent summaries of radionuclide field investigations conducted at the Nevada Test Site can be found in Nimz and Thompson (1992) and Smith (1998); older but equally valuable references are Buddemeier and Isherwood (1985), Buddemeier et al. (1991), and Borg et al., (1976).

### **QUALIFICATIONS**

While specific details for a representative number of sites will follow, synthesis of radionuclide data drawn from a wide variety of sources with sampling and analysis conducted by different organizations over many years is extremely difficult (Smith et al., 1995). Some of the qualifications that complicate the present data analysis:

 Most nuclear tests were conducted between ~ 500 and ~ 1200 m below the surface of the Nevada Test Site. Besides the obvious residual radiologic source term, the post-test environment is characterized by extreme heterogeneity due to the mechanical damage done to the rock units at the working (firing) point. For these reasons, the construction of wells and collection of water samples in a technically complex, expensive, and logistically complicated enterprise.

Selection of sampling sites was driven by differing priorities. In some cases, field-scale radionuclide experiments were deliberately conducted at preselected and optimized locations. In other cases the program exploited sites that were already targeted by defense programs for other subsurface

investigations.

 The phenomenology of underground nuclear explosions is complex and results in a heterogeneous distribution of radionuclides in the cavity and collapse chimney (Pawloski, 1999; Thompson, 1996). Because the residual radioactivity underground is highly fractionated, the resulting dissolved concentrations are non-uniform.

• An analysis of radionuclides collected from a single well does not represent an average for all cavity fluids. Radionuclide source terms are device specific

and cannot be averaged for the entire Nevada Test Site.

- Post-shot drilling conducted to collect solid samples for radiochemical diagnostics immediately after a test introduced large quantities of drilling muds, cements, or other additives into the cavity or near-cavity environment. Standard post-shot drilling procedure is to force drilling muds into the formation at the first sign of radioactivity to prevent radionuclides from circulating to the surface (LLNL, 1984). More than 13,000 barrels of bentonite and 7,500 barrels of sepiolite muds were introduced during drilling of post-shot holes by Los Alamos National laboratory (Bryant and Fabyrka-Martin, 1991). Bentonite clays and colloids present in the muds will sorb radionuclides; in addition these muds have high levels of natural radioactivity.
- Water samples are either collected using an evacuated two-liter bailer or by submersible or rotary pumps. Many hot wells particularly those that have been converted from post-shot entry holes consist of a single narrow (7.3 cm OD) carbon steel piezometer access tube, without a pump, completed in or adjacent to the cavity. Fluids trapped in an open piezometer tube will not circulate and adequate volumes of water cannot be produced to purge the well. Additionally raising and lowering a bailer in an open piezometer tube creates turbulence in the water column that compromises reproducible sampling. While bailers provide simple 'grab' samples of the fluid present in

the bore-hole, pumped samples are likely to be more representative of the bulk fluid composition in the near field.

Filtration and preservation treatments of water samples are variable. Work by Kersting et al. (1999) and Buddemeier and Hunt (1988) emphasize the importance of fine particulates (particles > 1.0 micron in size) and colloids (particles < 1.0 micron in size) to the transport of radionuclides which my sorb to these substrates. Filtered samples do not contain the particulate (retentate) fraction and may be lower in measured activity than their untreated equivalents. Acidified samples may not have the same major ion or radionuclide concentration as corresponding unacidified samples.</li>

While the results from recent field programs is considerable, the scope of this
effort should also be kept in perspective when trying to extrapolate
conclusions from approximately fifteen site-specific investigations to
comprehensive statement regarding the long-term effects of 828 underground
nuclear tests conducted at the Nevada Test Site (U.S. DOE/NV, 2000). These
fifteen sites constitute < 2 % of all the underground tests conducted at the
NTS.</li>

### SITE-SPECIFIC INVESTIGATIONS

Descriptions of selected near-field (hot well) sites are obtained from Buddemier and Isherwood (1985) who compiled much of this data in the mid-1980's. These data are augmented by HRMP and UGTA annual reports issued by LLNL and LANL (see Finnegan and Thompson, 2001; Thompson et al., 2000 and preceding documents). Radiochemical data has been compiled from a sub-set of the more than fifteen hot wells investigated at the NTS over the years. The purpose of this report is not to assemble all published hot well analyses but to evaluate a portion of the existing data to determine its suitability to define test and area specific hydrologic source terms. For this reason specific hot wells were selected for analysis where radionuclide data has been reliably collected over time and can be analyzed in the context of those radionuclides in solution, their maximum concentration, and the time-series evolution of this data. Time-series data is particularly important in determining controls on transport including radionuclide release, solubility, and sorption. In the following section, near field wells associated with the CAMBRIC, BILBY, DALHART, BASEBALL and INGOT tests on Yucca Flat and Frenchman Flat are discussed first followed by wells on Pahute Mesa associated with the CHESHIRE, ALMENDRO, and CAMEMBERT tests and the ER-20-5 site adjacent to the TYBO test.

### CAMBRIC (U-5e)

The CAMBRIC event has been extensively studied to determine the occurrence and distribution of radionuclides residual from a nuclear test as well as subsequent transport under both saturated and unsaturated conditions. Because it has been the focus of radionuclide migration studies since 1976, CAMBRIC is the most extensively studied of all the near-field sites on the Nevada Test Site. Complete descriptions of the radionuclide migration studies at CAMBRIC are contained in Hoffman et al. (1977) and Bryant (1992). CAMBRIC was also

recently the focus of a comprehensive effort to model the hydrologic flow and subsequent contaminant transport away from a nuclear test (Tompson et al., 1999) which required radiochemical data from the CAMBRIC wells to bound the numerical predictions as well as provide proper validation.

CAMBRIC was detonated on May 14, 1965 in emplacement hole U-5e. The announced yield of CAMBRIC was 750 tons (U.S. DOE/NV, 2000); in this respect, CAMBRIC is unusual due to its relatively low yield. The device was detonated in the alluvium 294 meters below ground surface and 74 m below the static water level. In 1974 RNM-1 was completed as a slant well within the original 1965 post-shot re-entry hole drilled for the collection of samples for radiochemical diagnostics. In addition to the collection of more sidewall core samples, RNM-1 was deepened for the collection of water samples from the cavity region. At the same time as RNM-1 was recompleted, RNM-2S was drilled vertically 91 m south of the U-5e emplacement and screened between 16 m and 41 m beneath the bottom of the CAMBRIC cavity. Approximately 500 m southeast of U-5e borehole, UE-5n was drilled vertically in 1976 to a total depth of 514 m immediately below the level of the water table. UE-5n was drilled initially to measure geologic site characteristics in central Frenchman Flat as well as monitor groundwater levels and the potential presence of contaminants. The experimental purpose of the RNM-1 and RNM-2S wells was to measure the distribution of radioactive materials in water and rock in the vicinity of a nuclear test and to conduct a field scale experiment involving radionuclide transport. The migration experiment entailed high volume pumping at RNM-2S to induce an artificial hydraulic gradient from the CAMBRIC cavity to the pumping well and documenting the subsequent transport of radionuclides away from the cavity.

The 1974 re-entry of RNM-1 into the cavity environment revealed that tritium and fission products from the test remained in the cavity/chimney complex. Radioactive species could not be detected in water pumped from a zone 50 m below the cavity. The tritium concentration in the chimney was ~ 2% of that in the cavity. <sup>239</sup>Pu and <sup>147</sup>Pm were largely retained in the cavity melt glass, while <sup>137</sup>Cs and <sup>144</sup>Ce had intermediate and <sup>90</sup>Sr, <sup>106</sup>Ru, <sup>125</sup>Sb had lower retention factors (relative to the residual CAMBRIC radionuclide source term). Finally, the ratio of <sup>85</sup>Kr to tritium in water samples ranged from 0.3 in the cavity to 1.5 in the chimney.

The pumping experiment at RNM-2S started and October, 1975 and ran nearly continuously until 1991. Pumps were initially run at 1100 liters per minute and increased to 2300 liters per minute before being shut-off. Two years after the pumping started, tritium appeared at RNM-2S after ~ 1.3 E+6 m³ of water had been pumped. Subsequently, low but steadily increasing concentrations of <sup>85</sup>Kr, <sup>36</sup>Cl, <sup>129</sup>I, <sup>106</sup>Ru and <sup>97</sup>Tc were measured in the water pumped from RNM-2S. Concentrations of these radionuclides rose sympathetically with tritium and reached a peak in 1981 before regularly declining. Cations including <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>239</sup>Pu were not detected in the water pumped from RNM-2S although, as mentioned above, they were detected in the water pumped from the

cavity. More than ~ 1.5 E+7 m³ of water was pumped from the RNM #2S by the end of the experiment reducing tritium concentrations in RNM #1 to below regulatory limits. As summarized by Nimz and Thompson (1992), the results of the CAMBRIC pumping experiment demonstrated that given a sufficient hydraulic gradient, groundwater will transport dissolved anions and noble gases that move conservatively with groundwater and do not sorb or only sorb weakly onto exposed rock and mineral surfaces. Radionuclides with high sorption coefficients and lower solubilities are not readily transported. Results from the CAMBRIC experiment are notable because of the unprecedented scope of this study. Accordingly these data and resulting interpretations form the basis for much of the understanding of field scale radionuclide migration at the Nevada Test Site and elsewhere. Data from RNM#1 and RNM-2S are presented in Tables 3 and 4 respectively.

UE-5n located 500 meters southeast of the U-5e emplacement hole exhibits regular increases in tritium concentration. Tritium has been measured over the past decade as part of a long-term radiological monitoring program. Although not tabulated, the tritium concentration has increased each year from 1989; rising from value of 17Bq/L in 1989 to 4.81E+3 Bq/L in when last sampled in 1999. Together with the rise in tritium, slightly elevated activities of <sup>36</sup>Cl, <sup>99</sup>Tc, and <sup>129</sup>I are detected in waters pumped from UE-5n with low volume Bennett pumps. Surprisingly <sup>14</sup>C is not elevated above natural background in these same samples. The source of the contaminants at UE-5n is not known with certainty. Initial modeling coupled with sampling at the UE-5n suggests that radionuclides are derived from water pumped from RNM-2 and allowed to infiltrate through the unsaturated zone along a kilometer-long ditch extending from the CAMBRIC site southeast to the Frenchman Lake playa (Tompson et al., 2002).

### BILBY (U-3cn)

The BILBY nuclear test was conducted on September 13, 1963 with an announced yield of 249 kilotons (U.S. DOE/NV, 2000). The test was the first conducted below the water table at the Nevada Test Site (Buddemeier and Isherwood, 1985) and was conducted in the central portion of Yucca Flat. The working point is in zeolitized, nonwelded Pre-Rainier Mesa tuffs that overlie the Paleozoic carbonate aquifer. The test was detonated at a depth of 713.2 m (2,340 feet). The pre-shot water table was at a depth of 502.9 m (1,650 feet). Because of BILBY's relatively large yield, and because the working point was separated by only 146 meters (481 feet) from the top of the Paleozoic carbonate aquifer, the BILBY test has been the focus of radionuclide studies for some time (U.S. DOE/NV, 1998; Nimz and Thompson, 1992). A review of field radionuclide experiments at BILBY is found in U.S. DOE/NV (1998), Buddemeier and Isherwood (1985), and Nimz and Thompson (1992).

The BILBY test is of interest to NTS radionuclide programs because it was one of the highest yield test conducted on Yucca Flat and was conducted below the water level proximal to the regional carbonate aquifer. Local pre-test hydrologic heads were disturbed for almost five years after the explosion. As summarized

by U.S. DOE/NV (1998), measurements of water levels in nearby wells immediately following the detonation indicated that water levels rose in response to a pressure pulse associated with displacement of rock around the working point. Hydraulic pressures also increased in the underlying Paleozoic sediments. Time-series water levels measured in the post-shot indicated that it took more than five years for the water level to recover to the pre-test level. These fluctuations suggest that the test had a profound influence of the hydrology of the site. Finally, because the test was conducted in 1963 and represents one of first and largest underground nuclear tests conducted beneath Yucca Flat, the hydrologic source term from this test is considerable and has now evolved for almost four decades. For this reason concentrations of radionuclides might represent levels anticipated under longer-term ambient hydrogeologic conditions.

The operational history of radionuclide migration studies at BILBY involve two borings: U-3cn PS#2 which was a post-shot re-entry hole and U-3cn #5 which was a satellite well drilled to the Paleozoic basement. The post-shot hole was drilled through the event chimney ten days after the test. The hole was drilled to at total depth of 793.4 m (2,603 feet) and, after sidewall samples were taken, the hole was cased. Soon after the casing was perforated, the casing collapsed at a depth of 587 m (1,925 feet) due to blocks shifting in the chimney rubble. The reentry well was open for sampling only in the interval of the event chimney where it was perforated from a depth of 512 to 527 m (1,679 to 1,729 feet) prior to a pump being set in the hole. The well was sampled intermittently from 1963 until 1984 when the pump malfunctioned. The U-3cn #5 was drilled in September, 1965 to a total depth of 923.5 m (3,030 feet) at a location 122 m (400.2 feet) southeast of U-3cn PS#2. This well was drilled to determine the depth of the Paleozoic rocks beneath BILBY and the distribution and intensity of radioactivity surrounding the emplacement hole. The well penetrated dolomitic limestone at a depth of 863.2 m (2,832 feet). It was cased and cemented so that the only water produced is from the Paleozoic section. At its closest, the hole is 47m from the outer edge of the cavity. Different pumps were set in the well and water was produced until the pump failed in 1981. The two BILBY wells were rehabilitated starting in December, 1996. The existing pump was replaced in U-3cn PS#2 and water pumped from the well in January, 1997. At U-3cn PS#2, a new pump was installed on carbon steel tubing with its intake at approximately the same depth (687.6 m or 2,256 feet) as the original pump. After the landing the new pump, water was produced from the well. Radiochemical data from U-3n PS#2 and #5 are compiled in Tables 5 and 6 respectively.

### DALHART (U-4u)

The DALHART test was conducted on October 13, 1988 in hole U-4u with an announced yield of less than 150 kilotons. The test was conducted at a depth of 640 meters (2100 feet) in Tertiary Tunnel Beds tuffs of Yucca Flat. The standing pre-shot water level was at a depth of 508 meters (1667 feet). Mathews et al. (1994) provides a complete description of the test and associated post-shot drilling and logging operations. A slant post-shot re-entry hole (U-4u PS 2a) was drilled into the chimney region of the test in 1990 and a 7.3 cm OD tube was

inserted which was subsequently perforated from 472 meters (1548 feet) to 501 meters (1643 feet) vertical depth (Thompson, 1999). Initial sampling in 2L bailers proved problematic due to the high concentration of particulates in these samples. In 1995, the chimney formation was pressurized to air-lift fluids from this well but this approach proved unsuccessful. Particulates again compromised samples produced from bailers in 1997. Using Bennett pumps working in tandem, 200 L of water was pumped from the post-shot well in 1998. A few cubic meters of water were also pumped from the well in 1999, however problems with the pumps and lines precluded extensive production. DALHART marked the first time Bennett pumps were successfully used in a contaminated near-field well to produce pumped samples which are more representative of the formation than those obtained using evacuated bailers. Radionuclide concentrations for DALHART are provided in Table 7.

### BASEBALL (U-7ba) and INGOT (U-2gg)

The BASEBALL and INGOT tests were conducted on January 15, 1981 and March 9, 1989 respectively in the northern half of Yucca Flat. The announced yield of both BASEBALL and INGOT was 20 to 150 kilotons (U.S. DOE/NV, 2000). The BASEBALL test has was conducted in hole U-7ba at a depth of 563.9 meters in the Tunnel Beds tuffs. The depth of the pre-shot static water level was 512 m. INGOT was emplaced in hole U-2gg at a depth of 500 m above the depth of the pre-shot water level at 565 m. INGOT was fired in the Timber Mountain Tuff. These two tests were drilled back many years after detonation for the purposes of developing near-field monitoring wells. Results of the analyses of solid samples provided valuable information on the migration of radionuclides by gaseous transport at the edge of a test cavity and the long-term behavior of radionuclides in a saturated test cavity (Thompson, 1996; Smith et al., 1996b; Smith, 1998).

The BASEBALL drill-back consisted of a slant main hole and a sidetrack completed at a slant depth of 711 m (or 665 m vertical depth) within the test cavity. Access was provided by 7 cm diameter carbon steel tube which was slotted below 600 m. The INGOT drill-back consisted of a slant hole that passed within 10 m of the edge of the explosion cavity and was completed at a slant depth of 726.5 m beneath the cavity and immediately below the level of the water table. At INGOT 14 cm diameter stainless steel casing was run in the hole with slotted completions below the water table. Pumps were slated to be installed in both holes to produce fluids representative of those within the cavity or near-cavity formation.

Water sampling at the two wells was compromised due to the inability of the pumps to return water to the surface. In the case of BASEBALL, the extensive use of drilling muds and residual particulates in the saturated cavity clogged both bailers and pumps. After several attempts to jet water in the hole to dilute and break-up the muds, further development attempts were abandoned. At INGOT, problems with casing separation doomed development of the well. While the submersible pump worked, water leaked out the tubulars and could not be collected at the well-head. Bailers were also deployed at INGOT and

water samples were collected and analyzed. These results are presented in Table 8.

### CHESHIRE (U-20n)

The CHESHIRE nuclear test was conducted on February 14, 1976 with a yield in the range of 200 to 500 kilotons (U.S. DOE/NV, 2000). The working point of the test is in the mafic poor Calico Hills formation which consists of a thick sequence of fractured rhyolite lava flows extruded from the Silent Canyon caldera on Pahute Mesa (Sawyer et al., 1999). The test was detonated at a depth of 1167m, and is well below pre-shot water table depth of 625m. A comprehensive review of field radionuclide experiments at CHESHIRE is provided by Buddemeier et al. (1991) and Sawyer et al. (1999). As noted by Buddemeier and Isherwood (1985), the CHESHIRE test is of interest to NTS radionuclide programs for several reasons:

 CHESHIRE is a larger yield test conducted deep below the water table in transmissive geologic units on Pahute Mesa. A number of the tests conducted prior to the 150 kiloton yield limit specified by the Threshold Test Ban Treaty on Pahute Mesa are – to first order – geologically and radiochemically similar, and results at CHESHIRE are generally applicable to these higher yield, below-water table detonations.

The CHESHIRE site is close to the northwestern edge of the Nevada Test Site
in rocks with high permeability; for this reason, characterizing the evolution
of the hydrologic source term in this part of the Nevada Test Site is important
to assessments of potential radionuclide migration beyond the boundaries of

the NTS.

• The CHESHIRE test represents the only location at the Nevada Test Site where both the radionuclide and hydrologic source term were measured immediately following a nuclear test within a well characterized hydrostratigraphic and fractured rock environment. Radiochemical data has been returned over a twenty-year interval. CHESHIRE represents the only "cradle to grave" nuclear test that can be studied to assess the long-term evolution of the hydrologic source term.

Initial studies suggest that relatively insoluble radionuclides sourced in the CHESHIRE test cavity may move conservatively by attachment to

groundwater colloids (Buddemeier and Hunt, 1988).

This site is being modeled as part of a large-scale effort to numerically predict
hydrologic flow and radionuclide transport from a underground nuclear test
in a system characterized both by thermal gradients, fracture flow, and
transient hydrology (see Pawloski et al., 2001 in review).

A brief history of operations at the CHESHIRE site is important to properly interpret the radiochemical data. The following is adapted from Buddemeier and Isherwood (1985). A re-entry hole (U-20n PS1 DD-H) was slant drilled during June and July, 1976. The hole was subsequently side-wall cored to a slant depth of 1378 m to recover glass and debris samples from the cavity region. Casing was perforated at the slant depth of 1322m, a pump was emplaced, and a

water sample was produced in September, 1976. This pump failed after only ~ 1500 gallons of water was produced. A bridge pump was then set above this perforated interval and the hole was logged again and re-perforated at a slant depth of 1293 to 1306 m. Samples were collected intermittently from this "cavity interval" from September, 1983 until May, 1985. At this time the slant hole was plugged again using a bridge plug and re-perforated approximately 250 m above the cavity horizon (812 to 913 m slant depth). Water was pumped from the "formation interval" from May to November, 1985. In June, 1987 a satellite well (UE-20n #1) was drilled approximately 300m downgradient from U-20n to a vertical depth of ~ 1000m to interrogate water flowing in a downgradient direction from the CHESHIRE formation interval; water was produced intermittently from this well in 1987 until the last sampling in July 1988. After pumping was suspended in the mid-1980's, the CHESHIRE formation interval was intermittently sampled by wire line bailer until the U-20n PS 1 DD-H well was rehabilitated in 1998. At that time, the casing was again perforated above the bridge plug in the formation interval at a depth of 812 to 913 m and water pumped from the hole; in September, 1998 the bridge plug was drilled out and the hole perforated in the cavity interval at a depth of 1250 to 1253 m slant depth before water was pumped from the level of the cavity. CHESHIRE cavity and formation concentrations data are listed in Tables 9 and 10 respectively. Data for the CHESHIRE satellite are provided in Table 11.

### ER-20-5

The ER-20-5 well cluster is located on Pahute Mesa in Area 20 in the northwestern corner of the NTS. The well cluster was constructed to characterize the nature and extent of radionuclide contamination from the TYBO (U-20y) nuclear test which was conducted on May 14, 1975 with an announced yield range of 200 to 1000 kilotons. The cluster was developed within ~ 300 m of the surface ground zero of the TYBO test. TYBO was detonated at a depth of 765 m (2,510 feet) and approximately 135 m (443 feet) below the static water level in hole U-20y. The working point of the test was located in zeolitic, nonwelded tuff of the Topopah Spring Tuff, which is the principal aquifer affected by the emplacement hole. TYBO was selected for near-field drilling because of the relatively large yield of the test, its hydrogeologic setting, the time elapsed since detonation, and the relatively shallow depth of burial.

Well cluster ER-20-5 consists of three bore-holes drilled on the same pad in late 1995. Well ER20-5 #1 was drilled to a depth of 860.5 m (2,823 feet) and was completed in a welded ash-flow tuff aquifer approximately two cavity radii southwest of the edge of the TYBO collapse chimney. Well ER20-5 #3 was drilled to a depth of 1,308.8 m (4,294 feet) and was completed in an underlying lava-flow aquifer of the Calico Hills formation. Well ER-20-5 #2 was abandoned due to drilling problems with no attempt to secure a completion. A comprehensive completion report for the ER-20-5 cluster is provided by the Department of Energy, Nevada Operations Office (U.S. DOE/NV, 1997c).

Water samples were pumped from this well in 1996, 1997, and 1998. A complete description of the well sampling and analysis program is provided by Kersting et

al. (1999). Radiochemical analysis of water samples collected from well ER-20-5 #1 indicated the presence of <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>155</sup>Eu in addition to tritium and other soluble fission products. The detection of europium is notable since rare-earth isotopes are relatively insoluble and are only infrequently encountered outside of the immediate cavity environment (Buddemeier and Hunt, 1988). Because these elements exhibit low solubilities, transport by means other than dissolved transport was suspected and water samples were sequentially filtered to determine if the radionuclides in the water were associated with colloids. Groundwater which had been pumped into 200L barrels was serial filtered using 1000 nm, 50 nm, and 10,000 nominal molecular weight (~ 7 nm) filters. The particulate material (> 1000 nm), the colloidal size fractions (between 1000 – 50 nm, 50 – 7 nm) and the ultrafiltrate or dissolved fraction (< ~ 7 nm) were collected and analyzed for 3H, gamma-emitting radionuclides, and plutonium isotopes.

<sup>3</sup>H, <sup>60</sup>Co, <sup>137</sup>Cs, <sup>152</sup>Eu, <sup>154</sup>Eu, <sup>155</sup>Eu and <sup>239+240</sup> Pu were detected in ER-20-5 #1 groundwaters and <sup>3</sup>H, <sup>60</sup>Co, <sup>137</sup>Cs, and <sup>239+240</sup> Pu were detected in ER-20-5 #3 groundwaters at concentrations 1000 to 20 times lower than the #1 well. Eu isotopes were not detected in the #3 well but may be below the analytical detection limit.

For the filtered samples, the tritium concentration was essentially unchanged passing through the filters. In contrast, greater than 99% of the europium and plutonium isotopes measured in groundwater from well #1 were associated with the colloid and particulate fractions. 91% of the <sup>60</sup>Co and 95% of the <sup>137</sup>Cs was associated with the particulate and colloid size fractions. High precision measurements of plutonium isotope ratios were performed in order to identify the source of the plutonium.

The consumption of nuclear fuels during a nuclear test imparts a unique <sup>240</sup>Pu/<sup>239</sup>Pu isotopic ratio that can be used to fingerprint the source of the plutonium measured wells ER-20-5 #1 and ER-20-5 #3. The <sup>240</sup>Pu/<sup>239</sup>Pu isotopic ratio of unfiltered groundwater in well #1 matches that measured in well #3 as well as the plutonium isotope signature of the colloidal fraction from well #1. In addition, all the plutonium isotope signatures match the BENHAM test but do not match the neighboring TYBO, MOLBO, or BELMONT tests or local NTS atmospheric fallout. The maximum concentration of plutonium measured was 2.3 E-2 Bq/L which is below federal drinking water standards. By determining the source of the plutonium, analyses of groundwater at the ER-20-5 cluster demonstrated that plutonium could be transported 1.3 kilometers from nuclear test where it originated. Additionally all the radionuclides, with the exception of tritium, were associated with particulates and colloids in the groundwater. X-ray diffraction and scanning electron microscopy indicated these solids consist of clays (illite and smectite), zeolites (mordenite and clinopilolite/heulandite) and cristobalite. For the first time, analysis of groundwaters from ER-20-5 implicated colloids in the transport of plutonium at ambient groundwater velocities through. fractured volcanic rock aquifers. Contrary to expectations, the source of the plutonium observed at ER-20-5 came from the BENHAM test over a kilometer

away and not the closer neighboring TYBO test. ER-20-5 #1 radionuclide concentration data is in Table 12; ER-20-5 #3 concentration data is in Table 13.

### **ALMENDRO (U-19v)**

The ALMENDRO test was conducted on June 6, 1973 with a yield in the range of 200 to 1000 kilotons (U.S. DOE/NV, 2000). The working point of the test is in the Crater Flat Tuff which consists of a thick sequence of bedded ash-low tuffs and rhyolite lavas extruded from the Silent Canyon caldera on Pahute Mesa (USGS, 1973). The test was detonated at a depth of 1063 m (3,487 feet) at a contact between the tuff (above) and rhyolite (below). The pre-shot water table was at a depth of 686.1 m (2,250 feet). The post-shot re-entry is designated U-19v PS-1DS and has been converted to a well for water sampling. Temperature logs run periodically into the ALMENDRO re-entry well starting in 1973 through the present indicate anomalously high temperatures persist in the test cavity. In 1996 a maximum temperature of ~ 157°C was recorded at a depth of 1147 m within the test cavity.

Rose et al. (2000a) noted that chemical and isotopic data for ALMENDRO groundwater samples collected within the past several years are not reflective of ambient groundwater conditions in central Pahute Mesa. For example, water samples collected in 1998 and 1999 show a significant positive shift in their  $\delta^{18}$ O isotope ratios relative to other deep Pahute Mesa groundwaters. These results are interpreted as evidence for high temperature water-rock isotopic exchange inside the ALMENDRO cavity. At temperatures between 100°C and 300°C, groundwater will readily exchange its oxygen isotopes with the surrounding rock. The high residual temperatures of ALMENDRO will promote isotopic exchange between the water and the rock, causing the water to become enriched in  $\delta^{18}$ O. In addition the dissolved inorganic carbon in water taken from the ALMENDRO cavity shows anomalous  $\delta^{13}$ C enriched carbon isotope signatures that may be the result of methanogenic processes involving the reduction of carbon dioxide (Rose et al., 2000a).

Immediately following the ALMENDRO test, the water level dropped ~ 250 m below the pre-test water level. Periodic monitoring of the water level in the post-shot re-entry well suggested that recovery of the water-table to pre-test levels took approximately twenty years. The anomalous water temperature, the unusual enrichment in oxygen and carbon isotope ratios, and the slow recovery of the water table to pre-shot levels suggests that water in the ALMENDRO cavity is only partially in contact with the ambient groundwater flow system in central Pahute Mesa. The chemical and physical measurements suggest the saturated ALMENDRO cavity represents a "quasi-closed" or partially isolated hydrologic system. The high temperatures and relative isolation of the ALMENDRO cavity was first recognized in 1973 and has persisted through time. The ALMENDRO test is important to studies of radionuclide migration at the Nevada Test Site because it demonstrates that under particular hydrogeologic settings involving particular pre-shot and phenomenological configurations, localized test-induced conditions may persist for more than three decades

following the detonation. In addition high temperatures encountered in the ALMENDRO cavity may dramatically alter the flow paths and flow velocities of water and the rates of reaction and affecting the solubility of radionuclides. The universal application of ambient groundwater temperatures and chemistry to the near-field environment at the Nevada Test Site is clearly inappropriate in some cases. Radionuclides in the ALMENDRO post-shot well are listed in Table 14.

### CAMEMBERT (U-19q)

The CAMEMBERT test was conducted on June 26, 1975 in hole U-19q with a announced yield in the range of 200 to 1000 kilotons (U.S. DOE/NV, 2000). The post-shot hole U-19q PS-1D was drilled into the test cavity which is estimated at a depth between 1400 and 1432 m. A liner was inserted in the hole to a depth of 1117m. In 1998 this hole was selected for rehabilitation based on the existence of the liner that extends almost to the cavity. During re-working the hole was found to be obstructed at 1121 m and a submersible pump was set at a depth of 916 m. During well purging, tritium concentrations were initially low (~ 3.7E+1 Bq/L) but soon stabilized at a concentration of 7.4E+5 Bq/L. In addition to 3H, 85Kr and 14C concentrations were among the highest of any reported concentrations of these isotopes on Pahute Mesa (85Kr: 4.0E+3 Bq/L and 14C: 107,004 percent modern carbon). <sup>36</sup>Cl was also elevated above environmental levels but not as severely as the <sup>85</sup>Kr and <sup>14</sup>C. No gamma emitting radionuclides other than naturally occurring species and <sup>137</sup>Cs were detected; the concentration of <sup>137</sup>Cs was 5.0E-1 Bq/L (Thompson, 2000).

A gas sample was also collected 107 m below the surface in the CAMEMBERT post-shot hole. The gas is representative of the vapor that remained undisturbed in the capped borehole (see Rose et al., 2000a for a complete discussion of the gas analysis). The gas is significantly depleted in oxygen relative to normal atmosphere and is enriched in hydrogen gas that characteristic of anoxic reducing condition favorable to methanogensis. The gas contains 1% methane and no CO<sub>2</sub>. Rose et al. (2000a) suggest that gas composition at CAMEMBERT evolved over time with consumption of O<sub>2</sub> and the production of CH<sub>4</sub> and H<sub>2</sub>. Microbial populations are most likely responsible for the methane detected in the gas sample.

The radiochemical and gas chemistry measured at CAMEMBERT is consistent with upwards radionuclide transport and quasi closed-system behavior previously encountered on Pahute Mesa. Conservative radionuclides (<sup>3</sup>H, <sup>14</sup>C, <sup>36</sup>Cl, <sup>85</sup>Kr) were able to ascend ~500 m upwards from the cavity region to the interval where groundwater was pumped. In addition, the CAMEMBERT cavity-chimney system was sufficiently isolated to preserve the non-equilibrium gas composition for more than 23 years since the time of the test.

### SIGNIFICANCE OF THE DATA

The following data analysis will concentrate of general conclusions about what radionuclide are observed in groundwater at the Nevada Test Site, their concentrations, times-series analysis of concentration data, and insights to

radionuclide transport at the NTS. One of the remaining questions is whether hot well data can provide information not only on the identity and concentration of radionuclides in the near-field but also on the controls of the solution and migration of radionuclides within and adjacent to the cavity-chimney complex. The justification for a rigorous program to regularly sample near-field wells can be summarized in several points.

First, measurement of radionuclides in the cavity and chimney provides data on which radionuclides are mobile, that can migrate from the cavity-chimney, and that contribute to the dose affecting potential down-gradient receptors. A test-specific and by-area inventory of more than forty long-lived radionuclides ( $t_{1/2} > 10$  years) has been prepared which provides an upper bound on the remaining atom abundance and activity in the subsurface at the NTS (Bowen et al., 2001). By comparing the atom abundance and activity of radionuclides in solution with those in the test-specific radionuclide inventory, a coarse estimate of solubility is possible. In addition, if the solubility is low, the data will provide assurance that radionuclide concentrations are below drinking water standards as well as designated contaminant dose boundaries. Contaminant concentrations in the cavity are expected to be highest and represent the most conservative measure of the dose. Assuming cavity concentrations do not exceed regulatory thresholds over time, there is some confidence that these levels will not be subsequently exceeded along a downgradient flow-path.

Second, analytical methodologies have improved dramatically over the past decade and provide a unique opportunity to measure the hydrologic source term at extremely low concentrations. <sup>3</sup>H, <sup>14</sup>C, <sup>36</sup>Cl, <sup>85</sup>Kr, <sup>90</sup>Sr, <sup>99</sup>Tc, <sup>129</sup>I, <sup>137</sup>Cs, <sup>235</sup>U, <sup>236</sup>U, <sup>237</sup>Np, <sup>239</sup>Pu, <sup>240</sup>Pu, and <sup>241</sup>Am may be measured at concentrations less than 0.05 Bq/L with high precision, high sensitivity mass spectrometry. In particular the arrival of the next generation multi-collector high-resolution inductively coupled mass spectrometry affords precise and accurate determination of isotope ratios to 0.01% with a detection limit of 2 E-15 grams for uranium and plutonium. Samples may be run with only simplified purification chemistry with a short analytical cycle per sample (10 to 20 minutes) which promotes through-put and cost-savings. These analytical advances enhance the capability of measuring a variety of radionuclides at low concentrations that otherwise were complicated to analyze, or were below method detection limits. For this reason state-of-theart analytical tools best exploit near-field sampling opportunities and provide a true and reliable measure of the hydrologic source term.

Third, data from near-field wells not only contains information on dissolved and suspended radionuclides in solution but also may be utilized to determine the environmental conditions bearing on the source and evolution of the water matrix containing radionuclides. In particular, temperature data, pH, bulk gas, major cations and anions, and stable (non-radiogenic) isotopes may be indicative of water that is not representative of ambient hydrologic conditions that existed prior to testing. As noted above, the temperature anomalies at ALMENDRO suggest the cavity has not lost significant amounts of heat in the nearly three decades since the time of test and has sealed itself from extensive communication from the surrounding aquifer (Rose et al., 2000a). Temperature profiles have also

proven to be important in understanding the transport pathways in the cavity and chimney environment; buoyancy effects may create vertical gradients through the permeable chimney which allow conservative radionuclides to ascend before being transported laterally in shallower aquifers (Maxwell et al., 2000). The vertical transport of radionuclides upwards from the cavity through chimney and subsequently down-gradient through shallower transmissive units is suspected to be the result of residual thermal effects at the CHESHIRE site as well as other deeply buried tests of Pahute Mesa (Sawyer et al., 1999). Data required for these interpretations is only obtained by regular monitoring of field parameters and associated water chemistry.

Fourth, regular sampling of hot-wells provides necessary experience in collecting radiologically contaminated samples, assessing their quality, trending the results, and establishing a radiological baseline which service both operational and regulatory needs. The ability to routinely return tritiated fluids from pumped or bailed holes requires specialized knowledge and training that is unique to sampling in the near-field. Routinely exercising the capability to return and analyze samples from the field enhances data quality objectives. Data returned from current and future hot-well campaigns can be added to the existing data-set to establish a radiological profile for each sampling site beneficial for a monitoring program to assesses changes in source-term model input parameters.

Finally, hot well data provide the only means to validate predictions of radionuclide concentrations in the near-field. LLNL has been developing hydrologic source term calculations in the near-field at the scale of an individual nuclear test (e.g., Tompson et al., 1999, Pawloski et al., 2001) which rely on measurements of radionuclides in groundwater in part to define the radiolgic source term to model. The resulting radionuclide flux can be checked by comparing model results with concentration data over time. On a test specific basis this comparison will by necessity be classified. Indeed LLNL will pursue just a comparison for its next phase of modeling the source tem at the CHESHIRE test on Pahute Mesa.

### PRIOR DATA ANALYSIS

Hot-well well data have been regularly published through the annual laboratory and field studies report related to radionuclide migration at the Nevada Test Site prepared by Los Alamos National Laboratory and Lawrence Livermore National Laboratory (e.g., Finnegan and Thompson, 2001; Thompson, 2000; Smith et al., 1999 and Smith et al., 1998 and prior reports). In addition summaries of this data have also been compiled and published but with less frequency (Buddemeier and Isherwood, 1985; Kersting, 1996). As noted above, comprehensive data analysis in these reports were intentionally abbreviated due to the complications inherent in year to year and well to well comparisons.

Recently, Finnegan and Thompson (2001) analyzed much of the data collected at the CAMBRIC, CHESHIRE, ALMENDRO and BILBY sites over the past three decades. Their primary conclusions are:

• A majority of the radionuclides detected at the CAMBRIC and CHESHIRE sites are volumetrically incorporated in the melt glass both at the time of the explosion and later. With the exception of tritium and <sup>85</sup>Kr, the radionuclide fraction dissolved in groundwater is 2 to 6 orders of magnitude less than in the solid phase (i.e., melt glass). These fractions do not appear to be perturbed through time (over more than twenty five years) evidenced by the extremely low concentrations of radionuclides measured in the cavity water up to the present (see Tables 3 and 9). The strong partitioning of radionuclides into the solid phase is observed consistently in the near-field sites investigated.

• Moving groundwater is required for radionuclides to migrate away from the cavity-chimney environment. This observation may be somewhat intuitive but is important for categorizing testing areas according to their potential for radionuclide movement. In areas with little or slow lateral groundwater flow such as Frenchman Flat or Yucca Flat, radionuclides detected in near-field wells by bailers often show concentrations gradients over a vertical interval that suggest the water in the saturated cavity-chimney environment is stagnant or is moving slowly. In contrast, on Pahute Mesa groundwater velocities are higher with average groundwater velocities ranging from 2 to 75 m/year (Laczniak et al. 1996). In the case of CHESHIRE and TYBO/BENHAM, radionuclides were encountered which had migrated significant distances (300 to 1300 m, respectively) from these sites due to prevailing hydraulic gradients encountered in the fractured rock aquifer.

 Some radionuclides are attached to colloids and particles that could be removed from water samples by filtration. This means of transport may be important on Pahute Mesa where ground water velocities are high and the load of ambient colloids consisting of clays, zeolites, quartz, and feldspars are approximately three orders of magnitude higher than in alluvial aquifers

(Finnegan and Thompson, 2001).

• At the ALMENDRO site high temperatures measured in waters produced from the cavity have persisted since the time of detonation. The higher temperatures might be expected to facilitate the leaching of the melt glass and exchange of volatile radionuclides and result in higher concentrations of radionuclides in solution. However, analyses of the water from the ALMENDRO cavity (see Table 14) reveal only trace amounts of ionic or refractory radionuclides. These results are difficult to reconcile with the temperature data and emphasize the need to better understand these closed-cavity chimney systems (Rose et al. 2000a).

Repeated water sampling in the carbonate aquifer at BILBY (U-3cn #5) indicates the absence of radioactivity. Because BILBY is one the few nuclear tests conducted at the NTS in the vicinity of the regional carbonate aquifer, potential releases from this test have significant environmental consequences. Although radionuclides have not been detected in well-U-3cn #5, the complex fracturing and solution permeability of the carbonate aquifer needs to be

better understood before the presence (or absence) of contamination in this portion of the carbonate aquifer is known with certainty.

### DATA ANALYSIS

The current data indicate a characteristic suite of radionuclides is commonly observed in groundwater near or adjacent to sites of expended nuclear tests. These data are important because they are a measure of anthropogenic radionuclides introduced to the groundwater regime by underground nuclear testing. However, analysis is not always straight-forward. In general, the hot wells described in this report were not designed for monitoring but rather were developed as observation wells to return data on radionuclide concentration and transport mechanisms. Well placement rather than radionuclide flux may be the reason that radionuclides have never been detected off the NTS. Similarly transport velocities should be considered as only minimum values. At the ER-20-5 and UE-20n#1 wells radionuclides are detected which originated from upgradient nuclear tests; <sup>3</sup>H/<sup>3</sup>He age dating provides a reliable measure of the time for the tritium to travel from the test to the contaminated well. However, contaminant plumes may extend for some distance beyond the well intercept implying more rapid transport velocities than calculated from the distance between the test and observation well only. The data evaluated here only allow for general statements regarding the radionuclide present in groundwater, their concentrations relative to the residual radiologic source term, and variations in concentration through time.

Radionuclides detected in Nevada Test Site waters correspond to longer-lived species ( $t_{1/2} > 1.0$  year) that have relatively higher solubilities or are associated with particles or colloids suspended in the groundwater. The expected radionuclide source term identified during initial studies of contaminant migration at the Nevada Test Site are still relevant to present studies. The fission product source term one half year, one year, ten years, and twenty five years after detonation of a 1 kiloton fission explosion from <sup>235</sup>U fuel is indicative of those radionuclides encountered in NTS near-field groundwaters (Borg et al., 1976).

Table 2 lists the activity of the radionuclide source term in TBq from fission and activation products residual from a 1 kiloton explosion burning <sup>235</sup>U with time elapsed since detonation (Borg et al., 1976).

TABLE 2

Activity (in TBq) with Time of the Radiologic Source Term from a 1 Kiloton Nuclear Detonation

Nuclide	Product	$t_{1/2}$ (years)	180 days	1 year	10 years	25 years
99Tc	Fission	2.13E+5	7.4E-4	7.4E-4	7.4E-4	7.4E-4
14C	Activation	5.73E+3	2.59E-3	2.59E-3	2.59E-3	2.59E-3
137Cs	Fission	30.07	6.6E0	6.6E0	5.2E0	3.7E0
90Sr	Fission	28.78	5.6E0	5.6E0	4.4E0	3.7E0
152Eu	Activation	1.35E+1	2.22E-1	2.15E-1	1.30E-1	5.19E-2
85Kr	Fission	10.76	8.1E-1	7.8E-1	4.4E-1	1.7E-1
60Co	Activation	5.27E0	4.4E-2	4.1E-2	1.11E-2	1.85E-3
125Sb	Fission	2.76	2.2E0	2.0E0	1.9E-1	3.7E-3
155Eu	Fission	4.71	1.1E0	9.3E-1	2.9E-2	7.4E-5
106Ru	Fission	1.02	1.4E+1	1.0E+1	1.9E-2	7.4E-7
54Mn	Activation	8.55E-1	6.67E-2	4.44E-2	2.22E-5	7.41E-11
144Ce	Fission	7.78E-1	1.33E+2	8.5E+1	2.8E-2	3.7E-8
95Zr	Fission	1.75E-1	1.62E+2	2.26E+1	3.7E-6	

In addition to the fission and activation products listed above, tritium residual from the consumption of thermonuclear fuels and actinides remaining from unburned primary fuel are present after a nuclear explosion. Mean activity values of the radiologic source term for <sup>3</sup>H from a total of 76 tests conducted on Pahute Mesa is 3.41E+4 TBq; the mean value for <sup>239</sup>Pu is 9.41E0 TBq and for <sup>235</sup>U (from the device) is 8.07E-4 TBq.

The longer-lived residual radionuclides listed above mirror those species detected in near-field groundwaters. In general the radionuclides can be broken into three groups: 1) those that are completely soluble in groundwater and 2) those that are soluble in groundwater but are ionic species that undergo ion exchange and sorption, and 3) those that are insoluble and associated with particulates or colloids. The first group includes <sup>3</sup>H, <sup>14</sup>C, <sup>36</sup>Cl, <sup>85</sup>Kr, <sup>99</sup>Tc, and <sup>129</sup>I. The second group includes <sup>22</sup>Na, <sup>54</sup>Mn, <sup>60</sup>Co, <sup>90</sup>Sr, <sup>95</sup>Zr, <sup>106</sup>Ru, <sup>125</sup>Sb, <sup>137</sup>Cs, and <sup>235</sup>U. The third group includes <sup>95</sup>Zr, <sup>144</sup>Ce, <sup>152</sup>Eu, <sup>154</sup>Eu, <sup>155</sup>Eu, and <sup>239</sup>Pu. These three groups form the basis for existing interpretations on the extent and controls on radionuclide migration at the Nevada Test Site.

## MECHANISMS OF RADIONUCLIDE TRANSPORT ELUCIDATED FROM NEAR-FIELD DATA

Radionuclides can be transported by processes resulting from the dynamics of the nuclear explosion and by longer term processes involving release to and transport by groundwater. The latter may include radionuclides released under saturated conditions and radionuclides transported by both gas and fluid flow in the unsaturated zone above the water table. Prompt transport occurs at timescales on the order of the nuclear detonation while transport involving

groundwater typically occurs over a scale of years to decades or longer. Both prompt and groundwater processes are effective in mobilizing radionuclides in the near-field and will be discussed separately.

### Prompt Process

Near-field drilling and hot well data clearly establishes the ability of the extreme temperatures and pressures associated with the explosion to rapidly move radionuclides into the accessible environment. At several sites, radionuclides are encountered in narrow intervals that - relative to thermodyamic or geochemical properties of the transported radionuclides - could not have been transported by any other means than dynamic processes. Nimz and Thompson (1992) and Pawloski (1999) provide an initial compilation of these observations and the associated tests where prompt injection has been reported. "Prompt injection" is a catch-all phrase for these dynamic, early-time processes. Although the mechanisms through which radionuclides can be transported outside the immediate cavity and chimney system is dependent on the geologic setting and phenomenology of each test, "prompt injection" broadly encompasses the movement of radionuclides as a plasma or high temperature gas along specific zones of weakness in the rock adjoining the explosion. Both high temperature refractory radionuclides (e.g., Rh, U, and Pu) and low temperature volatile radionuclides (e.g., Sr and Cs) are implicated. The production of significant amounts of incondensable gas (e.g.  $CO_2$ ,  $H_2$ ) by volatilization of carbonate minerals or the reduction of water can act as a carrier to rapidly move radionuclides upwards by displacement through the collapsed cavity and chimney or through fractures and porosity created in the formation adjacent to the blast. Gas in the cavity will ascend by in-fall of rubble from the overlying chimney and move gaseous radionuclides upwards in to chimney where low temperature radionuclides may condense on mineral and fracture surfaces.

Recent studies have implicated prompt injection in the transport of volatile and refractory radionuclides from the near-field to the far-field (Rose et al., 2000b). At the ER-20-6 site, three wells were drilled along a ~ 300 m radial line extending away from the BULLION (U-20bd) test. ER-20-6 #1 is located 166 m southwest of the surface ground zero of the BULLION test. After drilling ER-20-6 #1 a zone of intense radioactivity was observed in the profile for the spectral gamma log at a depth of 581 m in fractured volcanic rocks. Of note, this interval occurs 37 m above the static water level and radionuclides encountered here are thought to have been emplaced at the time of test. Laboratory measurements of gammaemitting radionuclides using fixed Ge(Li) detectors indicated the presence of <sup>22</sup>Na, <sup>125</sup>Sb, <sup>137</sup>Cs, and <sup>155</sup>Eu in bulk rock samples taken from this horizon. Secondary ion mass spectrometry (SIMS) was then used to anlayze radionuclides concentrations at ~ 60 micron diameter spots in polished rock samples. SIMS measurements detected the presence of <sup>22</sup>Na, <sup>137</sup>Cs, and a <sup>235</sup>U/<sup>238</sup>U isotope ratio enriched in <sup>235</sup>U relative to the natural isotopic abundance (0.02 measured relative the natural ratio of 0.007). In one instance all three species were found within a single 60 micron spot. The results indicate that radionuclides with ... highly contrasting volatilities and chemical properties (e.g., Na and U) may be deposited by prompt processes at the same location. The observation at ER-20-6

is consistent with the transport of radionuclides by "prompt injection" where a plasma produced by the explosion is forced outwards by the high pressures associated with the detonation along new or existing fractures adjacent to ground zero.

Radioactivity was encountered during drilling into the subsidence crater of the 1962 HYRAX nuclear test which was conducted in unconsolidated alluvium of Area 3 in Yucca Flat (Smith, 1998). Two vertical boreholes were drilled to a depth of 64 m before encountering a broad front of radioactivity. Radionuclides in core samples consisted of <sup>3</sup>H and <sup>137</sup>Cs. The <sup>137</sup>Cs is interpreted to have migrated upwards from the working point during cavity collapse and chimney in-fall. Over 90% of the  $^{137}$ Cs is produced from the  $\beta$ -decay of a gaseous  $^{137}$ Xe parent with a 229.2 second half-life. When the HYRAX cavity collapsed nearly 8 minutes after zero-time much of the <sup>137</sup>Cs chain yield was still in the form of <sup>137</sup>Xe. An abundance of calcite in the alluvium collected from the contaminated core samples (up to 50% of the fine grained soil fraction) implies significant fusion of carbonate at the time of the HYRAX detonation. The CO<sub>2</sub> gas liberated during the volatilization of the carbonate facilitated movement of the fission product gases in the <sup>137</sup>Cs chain. The <sup>137</sup>Xe gas precursor ascended upwards as the gas was displaced by debris that in-filled the cavity. The detection of <sup>137</sup>Cs at identical depths in two boreholes suggests the gas moved upwards along a broad front during the creation of the collapse chimney. Two neighboring tests, the 1962 BOBAC test and the 1963 JERBOA test, were conducted at similar depths in the same carbonate-rich alluvium with similar phenomenology but showed no evidence of <sup>137</sup>Cs ascent through the chimney. This implies that differences in radionuclide production, transport mechanisms, or collapse phenomena produced significantly different radionuclide distributions at BOBAC and JERBOA compared to HYRAX.

Gasous transport is further implicated in the observed distribution of radionuclides at the 1989 INGOT test (Smith et al., 1996b). Spectral gamma logging of a slant bore-hole that passed within ~ 10 m of the edge of the cavity encountered a discrete zone of radioactivity at the level of the working point which was immediately above the static water level. This interval is characterized by an abundance of hydrated smectites with > 20 weight percent  $H_2O$  which translates to a weaker rock strength. Analysis of sidewall cores indicated  $^{90}Sr$  and  $^{137}Cs$  were the only fission products present with an appreciable enrichment in  $^{137}Cs$  relative to  $^{90}Sr$ . The 32.3 second and 229.2 second half-lives of the parent isotopes of  $^{90}Kr$  and  $^{137}Xe$  allow these species to be transported as gases before decay to their longer-lived daughters. Rather than a pressure-temperature pulse that promptly injects condensing plasma, the results from INGOT suggest that some nuclear test cavities may be enveloped by an irregular perimeter of volatile radionuclides with gaseous precursors that were transported through fractures created or re-activated by the explosion.

The INGOT borehole was completed as a pumping well immediately below the puddle in the saturated zone. While water production proved troublesome, only dilute levels of radionuclides were encountered in samples taken 65 m beneath the test cavity. The ability to construct a well beneath a cavity is strategically

important to assess whether radionuclides leak from tests conducted at or immediately above the water table. The results from INGOT suggest only limited downward movement of soluble radionuclides from the test cavity to the underlying groundwaters.

Thompson (1996) analyzed a sequence of more than seventy sidewall-cores collected serially through the puddle and overlying collapse chimney of the 1981 BASEBALL test conducted in Yucca Flat. Despite the test being conducted below the water table and the glass and debris samples being saturated for more than 13 years, the radionuclide distribution and content was not disturbed. The persistence of the subtle variations in the radionuclide profile, including that for tritium, attests to the static nature of the cavity chimney environment in the absence of strong hydrologic gradients.

### Movement in Groundwater

### Yucca and Frenchman Flat

The hydrogeologic setting of Yucca and Frenchman Flats is an important factor in determining the long-term rate of radionuclide migration in areas extensively impacted by hundreds of underground nuclear tests. These basins containing thick sequences (up to ~ 600 m) of Quaternary aged alluvium that overlies a Tertiary volcanic sequence tuff, which in turn overlies Paleozoic carbonate rocks. Throughout much of Yucca Flat and Frenchman Flat the volcanic sequence functions as an aquitard, restricting vertical groundwater flow to the underlying carbonate aquifer. The carbonate aquifer is part of a laterally continuous regional flow system, and is therefore at greater risk in terms of contamination from nuclear testing. This aquifer is the only subsurface pathway by which groundwater exits these two basins (Laczniak et al., 1996). However, groundwater flow rates are slow; Winograd and Thordarson (1975) state that the hydraulic gradient is less than 0.1 to 4 meters per kilometer in the carbonate aquifer.

More than 600 of the 800 nuclear tests conducted at the Nevada Test Site were fired in the Frenchman Flat and Yucca Flat testing areas. Data from near-field wells in Yucca and Frenchman Flat suggests that the absence of an appreciable hydrologic gradient minimizes movement of radionuclides in the near-field. At CAMBRIC, BILBY, DALHART, and BASEBALL, independent lines of evidence together imply a lack of extensive radionuclide migration. At CAMBRIC, few radionuclides were initially measured outside of the cavity or in or adjacent to the chimney. 98% of the tritium was initially present in the cavity; concentrations were one or two orders of magnitude lower in the chimney and adjacent to chimney, and were at background below the cavity (Hoffman, 1979). The radionuclide migration pumping experiment demonstrated that at high pumping rates (1100 to 2300 liters per minute) radionuclides could be drawn out of the cavity and transported over a 91 meter interval to the pumping well. Transport data derived from the pumping trial indicates only highly soluble radionuclides including tritium, noble gases, anions, and oxyanions were mobilized. Tritium appeared at the RNM-2S well two years after the pumping

commenced in 1976 and concentrations of <sup>85</sup>Kr, <sup>36</sup>Cl, <sup>99</sup>Tc, <sup>106</sup>Ru, and <sup>129</sup>I rose with <sup>3</sup>H, reaching peak values in 1981. <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>155</sup>Eu, <sup>239</sup>Pu and other ionic species were not observed despite sixteen years of pumping. The time series data from the RNM-1 and RNM-2S wells confirms these trends. Of interest, the present concentrations of <sup>3</sup>H in RNM-1 are below the 740 Bq/L drinking water standard. It must be remembered that samples and analyses currently taken from the CAMBRIC system (i.e., RNM-1 and RNM-2S) are greatly disturbed due the pumping experiment and are not representative of an ambient system.

Time-series data from DALHART suggests that radionuclide concentrations may remain relatively constant due a lack of groundwater transport through the cavity-chimney system. Consistently high concentrations of radionuclides are produced more than ~ 100 m above the DALHART working point near the outside edge of the cavity. <sup>3</sup>H and <sup>85</sup>Kr are transported into the chimney while <sup>60</sup>Co and <sup>137</sup>Cs are detected only at very low concentrations. Like CAMBRIC it appears that only highly soluble radionuclides are transported beyond the initial confines of the DALHART cavity, whereas ionic species do not migrate appreciably.

Further evidence of limited migration of radionuclides under low hydrualic gradients is provided by the time-series analyses of water produced from the BILBY post-shot and satellite hole. BILBY is of importance to studies of radionuclide migration beneath Yucca Flat since pumped groundwaters have been returned intermittently since the time of the nuclear test in 1963. BILBY was one of the largest tests conducted beneath Yucca Flat (announced yield of 249 kilotons) depositing a substantive hydrologic source term populated by high concentrations of mobile radionuclide including <sup>3</sup>H, <sup>14</sup>C, <sup>36</sup>Cl, and <sup>85</sup>Kr (e.g., <sup>3</sup>H 3.25E+6 Bq/L in 1997). Despite the significant source term, the BILBY data provides indications that radionuclides have not migrated downward to the regional carbonate aquifer. The U-3cn #5 satellite well was completed in the carbonate aquifer 122m southeast and approximately 200 m deeper than the BILBY ground zero. After more than three decades of sampling, tritium levels measured in the satellite well are still five-orders-of-magnitude less than in the BILBY chimney. Refractory and reactive radionuclides have not been detected.

The BILBY results from the regional carbonate aquifer can be compared with results from historical sampling at the NASH (U-2ce) and BOURBON (U-7n) tests. The 1967 NASH test was conducted in Area 2 at a depth of 367.5 m immediately below the tuff-carbonate contact and above the static water table. At NASH, <sup>3</sup>H, <sup>35</sup>Kr, and <sup>22</sup>Na have been detected in a satellite well drilled 180 m from the test in the carbonate aquifer. The data show considerable variability over the 1978 to 1984 sampling window. It is suggested that either prompt injection or water level fluctuations caused the contamination to reach the satellite well. The 1967 BOURBON test was conducted at a depth of 560 m in silty limestone, 22 m above the static water level. At BOURBON, the nuclear test cavity likely intersected the water table. A satellite well drilled 137 m southeast of the BOURBON test was slotted to produce water from region affected by the lowermost cavity. Groundwater yields were extremely low which compromised sampling of representative formation waters. Low volume pumping produced

groundwater that was slightly enriched in tritium above regional background and contained no other detectable dissolved radionuclides. Continued pumping gave low but steadily increasing tritium concentrations. These results suggest that induced flow from pumping was necessary to mobilize the radionuclides, as observed at CAMBRIC.

### Pahute Mesa

In contrast with the low hydrologic gradients and groundwater velocities encountered in Yucca Flat and Frenchman Flat, Pahute Mesa is characterized by transmissive volcanic rock aquifers, steeper hydrologic gradients, and higher groundwater velocities. Although less than 100 nuclear tests were conducted on Pahute Mesa, many of these test had these high yields and detonated below the water table within rhyolite lavas and tuffs erupted from the Silent Canyon and Timber Mountain calderas. As noted by Bryant and Fabryka-Martin (1991) and Bowen et al. (2001), a majority of the groundwater accessible radionuclide inventory was deposited in Pahute Mesa. The combination of a large source term dominated by tritium, transmissive aquifers units, and steeper hydrologic gradients provides reason to responsibly assess the potential for radionuclide migration. Indeed, most cases of down-gradient transport of radionuclides have been reported on Pahute Mesa.

Pahute Mesa and Rainier Mesa lie within the southwest Nevada volcanic field and consist of Miocene rhyolites that were erupted from nearby calderas. The geology of Pahute Mesa consists of interbedded tuffs and lavas forming a volcanic stratigraphy in excess of 3000 meters thick. In general the lavas form aquifers due to the presence of interconnected and through-going fractures. Non-welded ash-flow and bedded tuffs are more likely to alter to zeolites which results in decreased permeability and development of confining units (Laczniak et al., 1996). Together the volcanic units in Pahute and Rainier Mesa make up a complex, three-dimensional interbedded framework of lava and welded tuff aquifers which are off-set by high-angle normal faults.

Recent observations from near-field wells drilled on Pahute Mesa have significant implications for understanding controls on radionuclide migration away from underground test sites. First, near-field data suggests that radionuclides thought to be relatively insoluble – including plutonium and rare earth elements - can be sorbed to mineral colloids and transported downgradient at ambient groundwater velocities. Second, soluble radionuclides may be encountered more than 500 meters above the working point either in the collapse chimney or within transmissive lava aquifers in hydrologic connection with the chimney. Down-hole thermal logs and hydrologic flow modeling implicates residual heat from the nuclear test results in buoyancy-driven groundwater transport up the chimney. Third, geochemical and down-hole thermal evidence suggests that some nuclear test cavities may not be in chemical or hydrologic connection with the ambient groundwater flow system. In some cases anomalous temperature and geochemical signatures in both the water and gas phase may persist for years post-test. Finally, in the few cases where representative pumped groundwaters were collected from the cavity

environment over several decades, concentrations of conservative radionuclides are decreasing due to dilution or dispersion while reactive radionuclides remain at constant concentrations or increase slightly due to continued reaction with the radiologic source term.

The results of water sampling at the ER-20-5 and CHESHIRE sites suggest that colloids play an important role in the transport of insoluble radionuclides. The suggestion of colloidal transport on Pahute Mesa is not new, having originated during sampling in the U-20n PS#1 DD-H and UE-20n #1 wells in the mid-1980's, (Buddemeier and Hunt, 1988). The initial work reported that nearly 100% of the transition elements (Mn, Co) and lanthanides (Ce, Eu) were associated with colloids of quartz and Ca-Na feldspars. Radionuclides sorbed to colloids were observed within the cavity, adjacent to the chimney 250 m above the working point, and 100 m laterally downgradient. The presence of radionuclides outside the cavity suggests that colloids served as the transport mechanism.

The detection of europium and plutonium at the ER-20-5 well cluster confirmed the ability of colloids to mobilize radionuclides and transport them considerable distances in groundwater at ambient groundwater velocities (Kersting et al., 1999). In the case of ER-20-5 radionuclides were transported more than 1.3 kilometers in less than thirty years. The actual transport distance may be greater - the plume may have traveled past the ER-20-5 well cluster - so this distance provides a minimum distance and conservative measure of transport velocities. Of note, like the initial findings at CHESHIRE, all the radioactivity with the exception of tritium was bound to particulates. Plutonium concentrations at ER-20-5 #1 were low and did not exceed 1E-14 M, and are identical to those measured in the CHESHIRE cavity. In addition, relative to ER-20-5 #3, higher concentrations of radionuclides were found in the shallower ER-20-5 #1 well that penetrates a fractured lava aquifer. Plutonium isotope signatures detected at the ER-20-5 well cluster were unambiguously traced to the BEHNAM test 1.3 kilometers up-gradient. This is surprising given the proximity of the TYBO test to the well cluster. There is no evidence that plutonium from TYBO has mixed with the plutonium encountered at the ER-20-5 well cluster.

At the ER-20-5, CHESHIRE, and CAMEMBERT sites on Pahute Mesa radionuclides in groundwaters are encountered appreciably above the working point of the tests. At ER-20-5 radionuclides were encountered 600 m above the BENHAM working point; at CHESHIRE radionuclides were encountered ~ 250 m above the working point; and at CAMEMBERT radionuclides were encountered 186 meters above the working point. In all cases soluble radionuclides including <sup>3</sup>H, <sup>14</sup>C, <sup>85</sup>Kr, <sup>36</sup>Cl are transported in high concentration to shallower levels. Concentrations of these species scale proportionally with tritium. Reactive radionuclides including <sup>60</sup>Co, <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>152</sup>Eu, <sup>154</sup>Eu, <sup>155</sup>Eu, and <sup>239</sup>Pu are detected at concentrations of half to two-orders-of magnitude less than cavity concentrations. As noted above, these species generally occur only in the presence of colloids. The conceptual model relies on residual heat from the explosion to heat water that has re-filled the cavity region to promote convective circulation of water to shallower intervals. In the case of BENHAM and CHESHIRE, the rising water encountered transmissive lava flow aquifers during

its ascent allowing radionuclides to be propagated in the direction of groundwater flow. Thermal logs taken from the CHESHIRE test were used to construct and bound comprehensive numerical models of flow in the near-field (Pawloski et al., 2001; Maxwell et al., 2000). Residual heat from larger yield tests like BENHAM with an announced yield of 1.15 Mt may considerably perturb the ambient groundwater flow system, accounting for the dispersal of radionuclides encountered in the ER-20-5 wells.

In other cases, thermal and geochemical effects observed in near-field wells suggest that some cavities are effectively isolated from the surrounding groundwater flow system. At ALMENDRO temperatures of 157°C were measured 23 years after the test. Rose et al. (2000a) suggest that the high temperatures may have remobilized silica sealing the cavity from extensive exchange with the surrounding groundwaters. Evidence that the cavity is isolated includes groundwater isotope enrichments that imply prolonged waterrock interaction at elevated temperatures and anomalous  $\delta^{13}$ C enriched carbon isotope signatures that may be the result of methanogenic processes involving the reduction of carbon dioxide (Rose et al., 2000a). Despite the high temperatures measured in the ALMENDRO cavity, ionic radionuclides (e.g., 137Cs) have only been measured at extremely low concentrations. Available evidence suggests the permeability of the surrounding rock may be a key factor governing the development of environmentally restricted conditions. Following the ALMENDRO test near-field water levels were depressed by 250 m relative to pre-test conditions, and required > 20 years to recover (Fenelon, 2000).

Analyses of gas samples taken from the CAMEMBERT borehole indicates the presence of hydrogen and methane, a depletion in oxygen indicative of anoxic, reducing conditions, and the absence of CO<sub>2</sub>. The gas compositions have persisted since CAMEMBERT was detonated in 1975, again possibly suggesting environmentally isolated conditions. Other sites with elevated down-hole temperatures include the FAULTLESS test conducted in 1968 at the Central Nevada Test Area (Hot Creek Valley, Nye County, Nevada); downhole temperature measurements in a post-shot hole drilled in the collapse crater are projected to be in excess of 100°C at a depths of 800 m or more.

Time-series interpretation of analyses of pumped water samples can be illustrative of controls on radionuclide concentrations in the near-field environment. Pumped water samples were collected from the CHESHIRE cavity from 1984 and again in 1998. Extensive pumping of cavity fluids provides reasonable assurance that the water samples are representative. Concentrations of radionuclides are reported as in Table 9 and are decay corrected back to the date of CHESHIRE test (February 1976) to permit comparison. Data are plotted in Figure 2 for <sup>3</sup>H, <sup>85</sup>Kr, <sup>99</sup>Tc, <sup>125</sup>Sb, <sup>129</sup>I, <sup>137</sup>Cs, <sup>239</sup>Pu, and <sup>241</sup>Am.

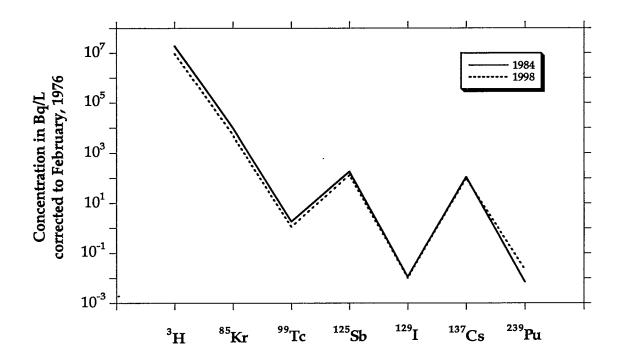


Figure 2. Plot of radionuclide concentration in Bq/L decay corrected to February, 1976 for radionuclides pumped from the CHESHIRE test cavity in 1984 and again in 1998.

<sup>3</sup>H, <sup>85</sup>Kr, <sup>99</sup>Tc, <sup>125</sup>Sb, slow slight decreases in concentration over the fourteen year interval while <sup>137</sup>Cs and <sup>239</sup>Pu remain constant or show slight increases in concentration. A first-order interpretation is that the soluble radionuclides are being diluted by groundwater entering the cavity environment or that radionuclides are dispersed as they migrate away from the cavity region, lowering the concentration. Sawyer et al. (1999) report that the effective volume in which tritium is distributed at CHESHIRE is about twice that of the cavity volume. This gave rise to concept of a "tritium exchange radius" where the distance of tritium deposition from the working point exceeds the measured cavity radius. As tritium continues to migrate out of the exchange zone, concentrations of soluble radionuclides in the cavity will decrease with time.

Alternately, with time, continued leaching of the melt glass and post-shot debris can release more volumetrically bound and surface deposited radionuclides from the cavity region to solution thereby increasing radionuclide concentrations. It should be emphasized that such time series analyses require pumped water samples produced from identical intervals regularly over time to permit comparison. To-date CHESHIRE is the only site that generates samples of the quality required for such an analysis.

### **CONCLUSIONS**

A regular program to collect groundwater samples from near-field wells and assess radionuclide migration processes is critical to the understanding of the long-term evolution of the hydrologic source term. Careful evaluation of these data is more imperative now than ever, as complex models are current being developed to describe the long-term evolution of the hydrologic source term (e.g., Tompson et al., 1999; Pawloski et al., 2001). Following is a paraphrased summary of a strategy document prepared providing the justification and importance this effort is as applicable today as it was when it was written almost two decades ago.

In response to needs to better manage resources that serve the defense programs mission at the Nevada Test Site as well as bound a predicted hydrologic source term from underground tests in support of the environmental management program, a program of sampling contaminated wells was initiated in the mid-1970's. An early intent of the effort was to "provide data and their interpretation which will quantify the potential for radionuclides to migrate away from underground nuclear explosion cavities and chimneys. If migration occurs, we can then determine the time-distance relationships of such migration under the conditions of a specific test. This information is applicable to the hydrologic studies related to long-term safety with regard to contamination of water supplies both on- and off-site. This information may also be useful to the National Nuclear Waste Terminal Storage (NWTS) Program with the deep geologic disposal of high-level radioactive waste." Futhermore, data collection needs were driven by need "to fulfill a moral obligation to preserve the safety of the public as a result of the weapons testing program which is a potentially hazardous activity. ...... The public must be protected from any unacceptable risks that have been conducted at the NTS. ...... Emphasis is on possible contamination of water supplies, which is potentially the greatest hazard. ..... We must demonstrate to a concerned and knowledgeable public that we can predict the behavior of radionuclides underground, whether they are from a nuclear test or from the disposal of nuclear waste." (RNM, 1983).

The near-field program has met part of these strategic objectives by identifying the radionuclides that occur with groundwater, and their concentrations. However, there has been a reluctance to over-interpret these data due to the factors outlined above (Smith et al., 1995). A significant amount of water chemistry data has been collected over several decades from near-field wells. However, data generated by this effort has not been evaluated comprehensively in the context of elucidating controls on the occurrence and mobility of

radionuclides in groundwater. The value of this time-series data is that it provides an empirical measure of the hydrologic source term. Individual processes that contribute to the presence of radionuclides in groundwater including leaching, precipitation, sorption, dilution, dispersion, and the presence of colloids may be revealed through trending of data collected over long periods of time. Using a conservative approach to assess the quality of this data and draw appropriate conclusions, near-field radiochemical concentrations provide empirical information required to effectively manage and remediate NTS groundwaters. Field based unique insight that otherwise might be missed through modeling or laboratory studies.

Three examples over the past decades emphasize the importance of a regular presence in the field to collect radiochemical data from near-field water samples. First, the unexpected presence of refractory, insoluble rare-earth fission and activation products in the CHESHIRE chimney provided indication that colloids were involved in radionuclide transport through fractured rock aquifers. Second, the detection of plutonium at the ER-20-5 well cluster that had originated more than 1.3 kilometers upgradient at the BENHAM test suggests that actinides can migrate at ambient groundwater velocities beneath Pahute Mesa. Third, the anomalously high temperatures recorded in the ALMENDRO cavity more than twenty years post event suggest that certain cavity systems may retain heat, and possibly radionuclides, for considerable times after the detonation. Each of these important observations would not have been predicted by conventional models or laboratory experiments without a near-field program to collect data on radionuclide concentrations.

In addition the near-field monitoring effort assumes increased importance by providing the only data critical to assessments of the long-term behavior of radionuclide migration away from expended tests. Although more than eight hundred underground tests were conducted at the Nevada Test Site, there are only approximately fifteen near-field study sites where water has been produced over the past thirty years for the analysis of radionuclides by the HRMP, UGTA or RREMP. Of note, only three of the fifteen study sites – CAMBRIC, CHESHIRE, and ALMENDRO – produce water from within a nuclear test cavity. For this reason, these data take on particular significance due to the relative scarcity of this information. The limited data-set also suggests that more insight is required from the cavity environment before representative statements can be made which apply to all of the Nevada Test Site. Ideally a comparative study which returns solids (i.e., melt glass and debris) and groundwater from identical intervals at the same time and place where the radiologic source term is also known would be optimal. To-date CAMBRIC and CHESHIRE are the only sites where radiologic and hydrologic source term data converge.

Despite the apparent gaps, data has been returned on an annual basis and provides a wealth of information to develop conceptual models of radionuclide migration at the Nevada Test Site as well as bound numerical predictions of the same.

Available evidence suggests that serious contamination of groundwater from the U.S. nuclear testing program is unlikely. Relative to the radiologic source term, only a small fraction of the actinides, fission products, and activation products are measured in near-field groundwaters. While the discovery of plutonium at the ER-20-5 wells was startling, the Federal drinking water standard for alphaemitting radionuclides in water is 15pCi/L or 75 times greater than the highest concentrations found at the well cluster (Federal Register, 2000). A combination of factors reduces the potential risk posed by these contaminants. A majority of the tests were conducted above the water table where radionuclides are assumed to be unavailable for transport by groundwater. Phenomenological effects concentrate actinides and fission products in the melt glass fraction where radionuclide release is governed by the dissolution rate of the glass. Melt glass dissolution does not appear to accelerated by radiation effects or high late-time temperatures. Some tests appear to seal themselves, resulting in little interaction with the ambient groundwater regime. A majority of tests conducted in Pahute Mesa, Yucca Flat, and Frenchman Flat were detonated in zeolitized ash-flow and ash-fall tuffs or alluvial sediments of the same that sequester radionuclides. Finally hydrologic gradients in the Frenchman Flat and Yucca Flat testing areas are low enough that lateral migration of radionuclides will be extremely slow and governed by flow rates of the regional carbonate aquifer, should radionuclides reach these depths. Where groundwater flow is static even tritium is relatively immobile.

#### A summary of the main points of this review:

- Soluble radionuclides include <sup>3</sup>H, <sup>14</sup>C, <sup>36</sup>Cl, <sup>85</sup>Kr, <sup>99</sup>Tc, and <sup>129</sup>I. These radionuclides are found as dissolved species and scale proportionally to concentrations of tritium (i.e., a 50% tritium concentration equates to a 50% concentration of <sup>85</sup>Kr when scaled to the cavity concentration of that species for a given nuclear test). Ionic radionuclides including <sup>90</sup>Sr, <sup>137</sup>Cs, <sup>152</sup>Eu, <sup>154</sup>Eu, and <sup>239</sup>Pu although encountered in cavity and chimney waters are not transported readily outside of this immediate environment.
- Moving groundwater is required to mobilize even highly soluble radionuclides; for this reason there is a higher potential for lateral movement of radionuclides on Pahute Mesa than Yucca Flat or Frenchman Flat.
- Time-series analyses provide information of the control of radionuclide migration including dilution, dispersion, leaching and precipitation.
- Relatively insoluble radionuclides including plutonium may be transported at ambient groundwater velocities through sorption to clay, zeolites, and feldspars colloids suspended in fractured rock aquifers. Resulting concentrations are below drinking water standards.
- Residual heat from underground nuclear tests may help mobilize radionuclides through the ascent of soluble species to more transmissive aquifers. Other tests have little interaction with ambient groundwaters and remain isolated for decades post-detonation.
- Prompt process may mobilize radionuclides along specific and narrow passages that are related the rock strength and geologic structure; gas phase

transport may cause volatile radionuclides to be deposited at shallower interval in a nuclear test chimney.

The value of the near-field well monitoring program cannot be underestimated for it provides the only empirical information on the concentration of radionuclides in and adjacent to the cavity-chimney environment. These data are necessary to the development of conceptual models of radionuclide migration and are needed to validate numerical predictions of the same. Time-series sampling and analysis of the groundwater from near-field provides insight into the evolution of the hydrologic source term. Only continued sampling of existing near-field wells and the strategic selection and development of new future sampling locations will ensure the data necessary to make credible and scientifically defensible statements regarding the migration of radionuclides at the Nevada Test Site.

#### **BIBLIOGRAPHY**

Borg, I.Y., Stone, R., Levy, H.B., and Ramspott, L.D., 1976, Information pertinent to the migration of radionuclides in ground water at the Nevada Test Site Part 1: review and analysis of existing information, Lawrence Livermore National Laboratory, UCRL-52078 Pt. 1, 216p.

Bowen, S., Finnegan, D.L., Thompson, J.L., Miller, C.M., Baca, P.L., Olivas, L.F., Geoffrion, C.G., Smith, D.K., Goishi, W., Esser, B.K., Meadows, J.W., Namboodiri, N., Wild, J.F., 2001, Nevada Test Site radionuclide inventory, 1951-1992, Los Alamos National Laboratory, LA-13859-MS, 28p.

Bryant, E.A., 1992, The CAMBRIC migration experiment – a summary report, Los Alamos National Laboratory, LA-12335-MS, 37p.

Bryant, E.A. and Fabryka-Martin, J., 1991, Survey of hazardous materials used in nuclear testing, Los Alamos National Laboratory, LA-12014-MS.

Buddemeier, R.W., Finkel, R.C., Marsh, K.V., Ruggieri, M.R., Rego, J.H., and Silva, R.J., 1991, Hydrology and radionuclide migration at the Nevada Test Site, Radiochimica Acta, v. 52/53, p. 275-282.

Buddemeier, R.W. and Isherwood, D., 1985, Radionuclide Migration Project 1984 Progress Report, Lawrence Livermore National Laboratory, UCRL-53628, 71p.

Buddemeier, R.W. and Hunt, J.R., 1988, Transport of colloidal contaminants in groundwater: radionuclide migration at the Nevada Test Site, Applied Geochemistry, v. 3, p. 535 – 548.

Farmer, D., Glew, T.R., and Faller, S.H., 1998, Measurement of current tritium concentrations in winter and summer precipitation at Las Vegas, Nevada (USA), Applied Sciences and the Environment, p. 97 – 112.

Federal Register, 2000, Environmental Protection Agency, Part II 40 CFR Parts 9, 141, and 142, December 7, 2000, v. 65, n. 236, p. 76708 – 76753.

Fenelon, J.M., 2000, Quality assurance and analysis of water levels in wells on Pahute Mesa and vicinity, Nevada Test Site, Nye County, Nevada, U.S Geological Survey, Water-Resources Investigation Report 00-4014, 68p.

Finnegan, D.L., and Thompson, J.L., 2001, Laboratory and field studies related to radionuclide migration at the Nevada Test Site in support of the Underground Test Area Program and Hydrologic Resources Management Project, Los Alamos National Laboratory, LA-13787-PR, 19p.

Hoffman, D.C., Stone, R., Dudley, W.W., 1977, Radioactivity in the underground environment of the CAMBRIC nuclear explosion at the Nevada Test Site, Los Alamos Scientific Laboratory, LA-6877-MS, 89p.

Hoffman, D.C., 1979, A field study of radionuclide migration, in Radioactive Waste in Geologic Storage, S. Fried, ed., American Chemical Society Symposium Series, n. 100, p. 149 - 166.

Kersting, A.B., 1996, The state of the hydrologic source term, Lawrence Livermore National Laboratory, UCRL-ID-126557, 30p.

Kersting, A.B., Efurd, D.W., Finnegan, D.L., Rokop, D.J., Smith, D.K., and Thompson, J.L., 1999, Migration of plutonium in groundwater at the Nevada Test Site, Nature, v. 397, p. 56-59.

Laczniak, R.J, Cole, J.C., Sawyer, D.A., Trudeau, D.A., 1996, Summary of hydrogeologic controls on ground-water flow at the Nevada Test Site, Nye County, Nevada, U.S. Geological Survey, Water Resources Investigations report 96-4109, 59p.

Lawrence Livermore National Laboratory, 1984, Post-shot drilling handbook, M.W. Butler, editor., M-148, 50p.

Mathews, M., Hahn, K., Thompson, J., Gadeken, L., and Madigan, W., 1994, Subsurface radionuclide investigation of a nuclear test, J. Applied Geophysics, v. 32, p. 279–291.

Maxwell, R.M., Tompson, A.F.B., Rambo, J.T., Carle, S.F., and Pawloski, G.A., 2000, Thermally induced groundwater flow resulting from an underground nuclear test, in Bentley, L.R. and others, eds., Proceedings of the XIII International Conference on Computational Methods in Water Resources, Calgary, Alberta, Canada, p. 45-50.

Nimz, G.J. and Thompson, J.L., 1992, Underground radionuclide migration at the Nevada Test Site, U.S. Department of Energy, Nevada Field Office, DOE/NV-346.

Pawloski, G.A., 1999, Development of phenomenological models of underground nuclear tests on Pahute Mesa, Nevada Test Site – BENHAM and TYBO, Lawrence Livermore National Laboratory, UCRL-ID-136003, in review, 49p.

Pawloski, G.A., Tompson, A.F.B., Carle, S.F., eds., 2001, Evaluation of the hydrologic source term from underground nuclear tests of Pahute Mesa at the Nevada Test Site: the CHESHIRE test, Lawrence Livermore National Laboratory, report in review.

Radionuclide Migration Program (RNM), 1983, Strategy document for the Radionuclide Migration Program, unpublished report available through the Analytical and Nuclear Chemistry Division HRMP document archives at Lawrence Livermore National Laboratory, 84p.

Rose, T.P., Pawloski, G.A., and Smith, D.K., 2000a, Environmentally closed nuclear test cavity-chimney systems: evidence and controlling factors, Lawrence Livermore National Laboratory, technical report in review, 31p.

Rose, T.P., Smith, D.K., and Phinney, D.L., 2000b, Secondary ion mass spectrometry measurements of volcanic tuffs containing radionuclides from underground nuclear tests, Radiochimica Acta, v. 88, p. 465-473.

Sawyer, D.A., Thompson, J.L., and Smith, D.K., 1999, The CHESHIRE migration experiment - a summary report, Los Alamos National Laboratory, LA-13555-MS, 32p.

Smith, D.K., Esser, B.K., Thompson, J.L., 1995, Uncertainties associated with the definition of a hydrologic source term for the Nevada Test Site, Lawrence Livermore National Laboratory, UCRL-ID-120322, 21p.

Smith, D.K., Esser, B.K., Kenneally, J.M., Nagle, R.J., Rego, J.H., and Royval, J.L., 1996a, Hydrologic Resources Management Program FY 1995 Progress Report, Lawrence Livermore National Laboratory, UCRL-ID-123486, 36p.

Smith, D.K., Nagle, R.J., and Kenneally, J.M., 1996b, Transport of gaseous fission products adjacent to an underground test cavity, Radiochimica Acta, v. 73.

Smith, D.K., Kersting, A.B., Kenneally, J.M., Rego, J.H., and Thompson, J.L., 1997, Hydrologic Resources Management Program FY 1996 Progress Report, Lawrence Livermore National Laboratory, UCRL-ID-126886, 40p.

Smith, D.K., 1998, A recent drilling program to investigate radionuclide migration at the Nevada Test Site, J. Radioanalytical and Nuclear Chemistry, v. 235, n. 1-2, p. 159-166.

Smith, D.K., Kersting, A.B., Rose, T.P., Kenneally, J.M., Hudson, G.B., Eaton, G.F., and Davisson, M.L., 1998, Hydrologic Resources Management Program and Underground Test Area FY 1997 Progress Report, Lawrence Livermore National Laboratory, UCRL-ID-130792, 92p.

Smith, D.K, Eaton, G.F., Rose, T.P., Kenneally, J.M., Hudson, G.B., Davisson, M.L., Benedict, F.C., and Criss, R.E., 1999, Hydrologic Resources Management Program and Underground Test Area FY 1998 Progress Report, Lawrence Livermore National Laboratory, UCRL-ID-135170, 76p.

State of Nevada, Nevada Department of Environmental Protection (NDEP), 1999a, Letter from P.J. Liebendorfer to R. Wycoff, Review of Corrective Action Investigation Plan, Revision I, Corrective Action Unit 98, Frenchman Flat, Federal Facility Agreement and Consent Order, September 21, 1999.

State of Nevada, Nevada Department of Environmental Protection (NDEP), 1999b, Letter from P.J. Liebendorfer to R. Wycoff, Review of Draft CAU 98 Frenchman Flat Groundwater Flow and Contaminant Transport Model and Source Term Modeling Document, November 18, 1999.

State of Nevada, Nevada Department of Environmental Protection (NDEP), 1999c, Letter from P.J. Liebendorfer to R. Wycoff, Review of Corrective Action Investigation Plan, Revision I, Corrective Action Units 101: Western and Central Pahute Mesa Nevada Test Site, Nevada Federal Facility and Consent Order, December 20, 1999

Thompson, J.L., 1996, Radionuclide distribution in a nuclear test cavity: the BASEBALL event, Radiochimica Acta, v. 72, p. 157-162.

Thompson, J.L., 1999, Laboratory and field studies related to radionuclide migration at the Nevada Test Site, Los Alamos National Laboratory, LA-13576-PR, 28p.

Thompson, J.L., 2000, Laboratory and field studies related to radionuclide migration at the Nevada Test Site, Los Alamos National Laboratory, LA-13701-PR, 23p.

Tompson, A.F.B., Bruton, C.J., Pawloski, G.A., eds., 1999, Evaluation of the hydrologic source term from underground nuclear tests in Frenchman Flat at the Nevada Test Site: the CAMBRIC test, Lawrence Livermore National Laboratory, UCRL-ID-132300, 319p.

Tompson, A.F.B., Smith, D.K., and Hudson, G.B., 2002, Analysis of radionuclide migration through a 200-m vadose zone following a 16-year infiltration event, Lawrence Livermore National Laboratory, UCRL-ID-146979, Preprint, Final LDRD Report, Tracking Code: 00-ERD-054, 27p.

- U.S. Department of Energy, Nevada Operations Office, 1993, Hydrology/Radionuclide Migration Program (HRMP) Redirection, Letter from J. Fiore to L. Smith, J. Magruder, and B. Church, February 24, 1993.
- U.S. Department of Energy, Nevada Operations Office, 1997a, Regional groundwater flow and tritium transport modeling and risk assessment of he Underground Test Area, Nevada Test Site, Nevada, DOE/NV-477, ITLV/10972-181, CD-ROM.
- U.S. Department of Energy, Nevada Operations Office, 1997b, Shaft and tunnel nuclear detonations at the Nevada Test Site: development of a primary database for the estimation of potential interactions with the regional groundwater system, DOE/NV-464, 62p.
- U.S. Department of Energy, Nevada Operations Office, 1997c, Completion report for the well cluster ER-20-5, DOE/NV-466.

- U.S. Department of Energy, Nevada Operations Office, 1998, Recompletion report for BILBY, DOE/NV 500.
- U. S. Department of Energy, Nevada Operations Office, 2000, United States Nuclear Tests, July 1945 through September 1992, DOE/NV-209. Rev. 15, 162p.
- U.S. Geological Survey, 1973, Geologic and hydrologic effects prediction and effects program, ALMENDRO (U19v) event, Pahute Mesa, Nevada Test Site, Special Studies 92, USGS-474-163, 38p.

Winograd, I.J., and Thordarson W., 1975, Hydrogeologic and hydrochemical framework, south-central Great Basin, Nevada-California, with Special Reference to the Nevada Test Site, U.S. Geological Survey Professional Paper 712-C, 126p.

# CAMBRIC (U-5e) RNM#1

All data is reported in Bq/L corrected to the date of the test (May 14, 1965). Note that the table does not attempt to tabulate every analysis made at RNM#1 and only provides representative analyses. NR= not reported; BDL = below detection limit.

Radionuclide	1979	1981	1982	1988	1990
-011	1 100 . 5	E 02E . 2	0.645.2	0.050.1	275.1
3H	1.17E+5	5.03E+3	2.64E+3	8.85E+1	3.7E+1
60Co	BDL	BDL	4.22E-3	NR	NR
106Ru	BDL	BDL	BDL	NR	NR
125Sb	1.46E-1	1.13E-1	BDL	NR	NR
137Cs	1.94E0	8.59E-1	8.56E-1	6.30E-1	3.48E-1
Comment	pump; unfiltered	pump; unfiltered	pump; unfiltered	pump; unfiltered	pump; unfiltered
Data Source	LLNL	LLNL	LLNL	LANL	LANL
Radionuclide	1991	1993			
3H	<3.7E+1	1.50E+2			
60Co	BDL	BDL			
106Ru	BDL	BDL			
125Sb	BDL	BDL			
137Cs	3.6E-1	4.85E-1			
Comment	pump; unfiltered	pump; unfiltered		`	
Data Source	LANL	LANL			

#### CAMBRIC (U-5e) RNM-2S

All data is reported in Bq/L corrected to the date of the test (May 14, 1965). Note that the table does not attempt to tabulate every analysis made at RNM#2S; in particular, time-series data for 85Kr, 36Cl, 99Tc, and 129I - which are known to mobile and have been detected in the RNM-2S well - are omitted (e.g., Bryant, 1992). However, the table provides representative analyses of tritium and many gamma-emitting radionuclides. NR= not reported; BDL = below detection limit.

Radionuclide	1978	1979	1980	1981	1982
3H	6.59E+4	9.63E+4	2.24E+5	2.21E+5	2.29E+5
106Ru	3.61E+1	3.38E+1	4.07E+1	BDL	BDL
125Sb	BDL	BDL	BDL	BDL	BDL
137Cs	9.22E-5	BDL	4.59E-4	6.88E-4	4.59E-4
Comment	pump; unfiltered	pump; unfiltered	pump; filtered	pump; filtered	pump; unfiltered
Data Source	LLNL	LLNL	LLNL	LLNL	LANL
Radionuclide	1984	1988	1991	1993	1999
3H	1.73E+5	8.07E+4	5.30E+4	4.56E+4	4.81E+4
60Co	BDL	NR	NR	BDL	NR
106Ru	BDL	NR	NR	BDL	NR
125Sb	BDL	NR	NR	BDL	NR
137Cs	6.89E-5	NR	NR	BDL	NR
		1.			
Comment	pump; filtered	pump; unfiltered	pump; unfiltered	pump; unfiltered	pump; unfiltered
Data Source	LLNL	LANL	LANL	LANL	LANL

TABLE 5

# BILBY (U-3cn PS#2)

All data is reported in Bq/L corrected to the date of the test (September 13, 1963). NR= not reported; BDL = below detection limit.

Radionuclide	1965	1977	1981	1985	1997
3H	1.00E+7	2.86E+6	4.74E+6	3.50E+6	3.25E+6
14C	NR	NR	NR	NR	6.41E0
36Cl	NR	NR	NR	NR	1.11E-2
60Co	9.41E+0	7.19E-2	3.17E-3	NR	NR
85Kr	NR	NR	NR	NR	2.67E+5
106Ru	4.15E+2	BDL	BDL	NR	NR
125Sb	3.42E-1	1.08E0	3.42E-1	NR	NR
137Cs	1.89E-1	1.66E-1	8.30E-2	NR	8.85E-2
144Ce	5.63E+0	NR	NR	NR ·	NR
239Pu	NR	3.17E-4	3.34E-5	NR	NR
Comment	pump; filtered	pump; unfiltered	pump; filtered		pump; unfiltered
Data Source	USGS	LLNL	LLNL	LLNL	LLNL & LANL

TABLE 6

# BILBY (U-3cn #5)

All data is reported in Bq/L corrected to the date of the test (September 13, 1963). NR= not reported; BDL = below detection limit.

Radionuclide	1980	1981	1997
3H	1.01E0	2.99E-1	1.99E+1
14C	NR	NR	4.07E-2
36Cl	NR	NR	1.48E-5
60Co	BDL	BDL	BDL
85Kr	NR	NR	BDL
106Ru	BDL	BDL	BDL
125Sb	BDL	BDL	BDL
137Cs	1.38E-4	BDL	BDL
239Pu	NR	NR	NR
Comment	pump; filtered	pump; filtered	pump; unfiltered
Data Source	LLNL	LLNL	LLNL & LANL

# DALHART (U-4u PS2A)

All data is reported in Bq/L corrected to the date of the DALHART test (October 13, 1988). NR= not reported; BDL = below detection limit.

Radionuclide	July 1997	July 1997	September 1998	August 1999
3H	9.7E+5	9.9E+5	1.40E+6	1.00E+6
60Co	NR	NR	1.0E0	5.8E-1
85Kr	3.2E+2	4.6E+2	NR	NR
125Sb	NR	NR	3.3E+1	2.4E+1
137Cs	NR	NR	3.3E0	2.0E0
Comment	bailed; unfiltered	bailed; unfiltered	pump; unfiltered	pump; unfiltered
Data Source	LANL	LANL	LANL	LANL

# INGOT (U-2gg PSE3A)

All data is reported in Bq/L corrected to the date of the INGOT test (March 9, 1989). NR= not reported; BDL = below detection limit.

Radionuclide	September 1994	September 1994
3H	3.28E+2	2.76E+2
85Kr	NR	5.30E-1
137Cs	3.78E-2	1.15E-1
Comment	bailed; unfiltered	bailed; unfiltered
Data Source	LLNL	LLNL & LANL

#### CHESHIRE (U-20n PS1 DD-H Cavity)

Data from the U-20n PS1 DD-H "cavity interval" within the test cavity. All data are reported in Bq/L corrected to the date of the test (February 14, 1976). Not every radionuclide analyzed from these samples is reported. In some cases, duplicate analyses were reported within a single sampling year; for instance LLNL and LANL each reported their own analyses for a particular sampling event. To facilitate comparison duplicate analyses were averaged if the variation between analyses was within one order of magnitude; if the variation exceeded an order of magnitude both values are listed. NR= not reported; BDL = below detection limit.

Radionuclide	1976	1983	1984	1998
3H	1.0E+7	2.3E+7	1.9E+7	9.4E+6
14C	NR	NR	NR	7.8E0
22Na	NR	2.8E-1	3.0E-1	NR
36Cl	NR	NR	NR	1.8E-2
54Mn	BDL	4.4E0	3.6E0	NR
60Co	BDL	2.0E-1	1.3E-1/5.2E-3	2.2E-1
85Kr	NR	NR	1.0E+4	5.2E+3
99Tc	NR	1.8E0	1.8E0	1.1E0
106Ru	1.0E+2	4.6E+2	4.2E+2	NR
125Sb	1.0E+2	2.0E+2	1.8E+2	1.3E+2
129I	NR	NR	1.1E-2	1.0E-2
134Cs	NR	8.1E-1	1.0E0	NR
137Cs	BDL	1.2E+2	1.1E+2	9.6E+1
144Ce	BDL	4.4E-1	3.7E-1	NR
152Eu	NR	3.5E-1	2.8E-1/4.0E-3	1.5E-1
154Eu	NR	6.1E-1	4.9E-1/2.0E-2	1.6E-1
155Eu	NR	1.5E0	1.4E0	NR
239Pu	NR	3.1E-2	7.0E-3	2.1E-2
241Am	NR	NR	NR	4.0E-3
Comment	pump; filtered	pump; filtered	pump; unfiltered	pump; unfiltered
Data Source	LLNL	LLNL & LANL	LLNL & LANL	LLNL & LANL

TABLE 10

# CHESHIRE (U-20n PS1 DD-H Formation)

Data from the U-20n PS1 DD-H "formation interval" above the test cavity. All data are reported in Bq/L corrected to the date of the test (February 14, 1976). NR= not reported; BDL = below detection limit.

Radionuclide	1985	1998
3H	1.6E+7	9.6E+3
14C 22Na	NR 4.7E-2	4.2E0 NR
36Cl 54Mn	NR 2.6E-1	9.1E-3 NR
60Co	2.9E-3	NR
85Kr 99Tc	1.5E+7 1.4E0	2.5E+3 7.4E-1
106Ru 125Sb	8.3E+1 1.5E+2	NR 5.5E0
1291	8.6E-3	4.9E-3
134Cs 137Cs	4.9E-2 2.4E+1	NR 2.6E-1
144Ce 152Eu	BDL 4.7E-3	NR NR
154Eu	8.9E-3	NR
155Eu 239Pu	2.6E-2   NR	NR 1.4E-4
241Am	NR	NR
Comment	pump; filtered	pump; unfiltered
Data Source	LLNL	LLNL & LANL

TABLE 11

# CHESHIRE (U-20n) UE-20n #1 Satellite

All data is reported in Bq/L corrected to the date of the test (February 14, 1976). NR= not reported; BDL = below detection limit.

Radionuclide	1987
3H	2.1E+7
14C	NR
22Na	1.5E0
36Cl	NR
54Mn	BDL
60Co	BDL
85Kr	BDL
99Tc	NR
106Ru	BDL
125Sb	3.6E+1
129I	NR
134Cs	NR
137Cs	4.4E-2
144Ce	BDL
152Eu	BDL
154Eu	BDL
155Eu	BDL
239Pu	NR
241Am	NR
Comment	pump; unfiltered
Data Source	LLNL

#### ER-20-5 #1

All data is reported in Bq/L corrected to the date of the TYBO test (May 14, 1975). NR= not reported; BDL = below detection limit.

Radionuclide	January 1996	June 1996	April 1997	July 1998
3H	2.11E+8	2.23E+8	8.80E+6	8.63E+6
14C	NR	2.3	NR	6.63
36Cl	NR	1.27E-1	NR	1.23E-1
60Co	1.08E0	3.0E+1	1.13E0	1E0
85Kr	NR	1.07E+3	NR	8.12E+1
137Cs	7.1E-1	9.34E-1	9.6E-1	8.33E-1
152Eu	1.68E-1	1.67E-1	1.6E-1	1.59E-1
154Eu	3.8E-1	3.6E-1	3.5E-1	3.52E-1
155Eu	3.6E-1	3.6E-1	2.9E-1	3.19E-1
239+240Pu	1.96E-2	9.63E-3	2.3E-2	NR
Comment	pump;	pump;	pump;	pump;
	unfiltered	unfiltered	unfiltered	unfiltered
Data Source	LLNL &	LLNL &	LLNL &	LLNL &
Data boute	LANL	LANL	LANL	LANL

TABLE 13

#### ER-20-5 #3

All data is reported in Bq/L corrected to the date of the TYBO test (May 14, 1975). NR= not reported; BDL = below detection limit.

Radionuclide	February 1996	July 1996	April 1997	July 1998
3H	7.8E+3	1.9E+4	1.81E+4	1.87E+4
14C	NR	7.03E-2	NR	6.41E-2
36CI	NR	3.7E-4	NR	4.1E-4
60Co	3.0E-2	6.0E-2	1E0	NR
85Kr	NR	NR	NR	NR
137Cs	1.04E-2	8.7E-3	1E-1	NR
239+240Pu	NR	3.0E-4	NR	NR
Comment	pump; unfiltered	pump; unfiltered	pump; unfiltered	pump; unfiltered
Data Source	LLNL & LANL	LLNL & LANL	LLNL & LANL	LLNL & LANL

TABLE 14

#### ALMENDRO (U-19v PS#1 DS)

All data is reported in Bq/L corrected to the date of the test (June 6, 1973). NR= not reported; BDL = below detection limit.

Radionuclide	1993	1993	1996	1996	1998	1999
3H 36Cl 85Kr 137Cs	8.5E+6 NR NR BDL	2.7E+7 NR NR NR BDL	NR NR NR 1.15E0	2.8E+7 NR NR NR NR	2.8E+7 NR 1.0E+4 2.3E-2	2.3E+7 7.9E-2 NR NR
Temperature (°C)	,		43	104		
Comment .	bailed; 2464' measured depth. 0.2 µm filtered	bailed; 3300' measured depth. 0.2 µm filtered	bailed; 3300' measured depth.	bailed; 3585' measured depth.	bailed; 3574' measured depth.	bailed; 3202' measured depth.
Data Source	LLNL	LLNL	LLNL	LLNL	LANL& LLNL	LANL

Radionuclide concentrations are remarkably low for samples produced from a saturated nuclear test cavity. However, metals have been detected in the ALMENDRO cavity water in addition to radionuclides. Recent analyses (LLNL-CES, 1999) of heavy metals in waters collected from 2997' measured depth in the U-19v PS#1DS hole indicate a Pb concentration in unfiltered water samples of 63  $\mu g/L$ , a Fe concentration of 12000  $\mu g/L$ , and an As concentration of 1716  $\mu g/L$ . ALMENDRO is only one of few near-field wells where heavy metals have been identified in water samples.